Valorization of beetle infected spruce to produce textile fibers and biofuels: Environmental sustainability evaluated by life cycle assessment

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ABSTRACT

To achieve a viable forest-based biorefinery, both the carbohydrate and lignin parts of the raw material should be valorized. While lignin-first approaches have successfully been applied to hardwoods, where up to 50% of the lignin –close to the 'theoretical maximum yield'– has been transformed to valuable monophenols; limited studies have targeted softwoods. Softwood lignin comprises lower amount of beta-ether bonds and this results in lower theoretical and observed yields of monophenols in reductive catalytic fractionation (RCF): below 5 wt% yield of initial biomass has been reported. In this study, we use beetle infected spruce, a softwood, as raw material. A fast fractionation was developed to give a pulp and a lignin fraction in the absence of transition metal catalysts. The carbohydrate matrix was valorized to dissolving grade pulp in 37 wt% from biomass (86% yield), and successfully spun to Lyocell fibers. The lignin fraction was dissolved in furfural –operating as green 'solubility-enhancing-agent'– to blend lignin in inert carrier liquids to promote controlled hydrotreatment to yield biofuels in 10 wt% (60% carbon yield) from initial biomass. Life cycle assessment (LCA) of the value-chain showed improved sustainability in several footprint categories compared to cotton production. Thus, upgrading of a considered forestry waste to high value textile fibers and biofuels has been achieved: in case of lignin beyond the 'theoretical maximum yield'. This is an important step to mitigate a future growing demand of textiles without negatively affecting irrigation or land use.

1. Introduction

Population growth and exhaustion of natural resources are counteracting challenges for this century [1,2]. By 2060, the world population is predicted to have grown from today's 8 billion to 10 billion [3]; crude oil, our main source for chemicals, materials and transportation fuel is forecasted to be nearly depleted [4,5]. To meet these counteracting challenges without deforestation, it is crucial to use current resources better, even considered wastes; substitute production of crops that compete with food production such as first generation biofuels and cotton [6,7].

The tree-killing bark beetles [8] cause damage to forests all around the world [9]. Softwoods [10], such as spruce and pine are vulnerable to bark beetles [11]. Due to climate change and monoclonal forests, the situation has worsened. When the climate becomes warmer and drier, the tree's natural defense towards bark beetles is weakened [12–14]. Between 2017 and 2019, over 270 million m^3 of standing timber only in Central Europe has been damaged primarily due to the spruce bark beetle [11]. Also elsewhere, as in the Americas, bark beetles have become a growing problem [15].

The beetles infect the tree and lay eggs that hatch after around 5 weeks. After maturing for around 3–5 additional weeks, the beetles

leave the tree, after which the tree dies. Removing infected trees have been proven to be the most efficient counter-measure to prevent spreading of the bark beetle. The massive amounts of biomass from beetle infected spruce is rejected from saw mills and kraft pulping and is currently incinerated to a low value. Due to high moisture content, the beetle infected spruce has a as low higher heating value (HHV) of around 17 MJ·kg^{-1} [16]. Taking into account the large volumes ∼ 100 million m^3 per year of damaged wood only in Europe it would be worthwhile to find technologies to upgrade this waste stream. If the infected wood could be upgraded to dissolving pulp for textile production, it would be an incitement to harvest these volumes and then benefits to reclaim areal for food production would be possible [17].

Current pulping technologies are focused on production of pulp using virgin wood. In practice, <50% of the wood is converted to pulp and the rest of the wood, mainly lignin, is incinerated to a low value [18]. Lignin comprises 20–30 wt% of the wood, however almost half of the energy content due to its higher calorific value (23–26 MJ/kg) as compared to carbohydrates (18 MJ/kg) [19]. Lignin is an aromatic polymer that could be a useful source of aromatics both for petrochemical and fuel applications where it would substitute petroleum-derived aromatics. From petrochemical perspective, the production of benzene, toluene and xylene that are important petrochemical building

blocks can be envisaged. From transportation fuels perspective, the direct production of aromatics from biomass-derived lignin is highly desirable as there is not any other source of aromatics in the natural resources. The aromatics play a vital role particularly in the gasoline pool due to their high octane numbers and current gasoline contains up to 35 vol% of aromatics according to the EU standard EN228. Aromatics are also very important for aviation fuels as they ensure good compatibility between the used engine sealings and kerosene, thus preventing fuel leakages. However, the traditional pulping processes resulting in sulfur-containing lignins have prevented these uses up to now, which could change with the advent of novel wood fractionation technologies.

Lignin-first approaches have emerged the last decade as an alternative to conventional pulping, where lignin valorization is considered in the design phase of fractionation [20–24]. State-of-the-art methodologies such as reductive catalytic fractionation (RCF) have demonstrated that near 'theoretical maximum yields' of monophenols can be obtained when transition metal catalysis is employed. The 'theoretical maximum yield' of lignin monomers is calculated as x^2, where x is the β-ether content in lignin (Fig. 1A) [25,26]. Hardwood lignin contains up to 70% β-ethers and experimentally 50 wt% yields of monophenols have been reported in RCF experiments, corresponding to 100% of the 'theoretical maximum yield' [27]. While there are numerous studies of lignin-first
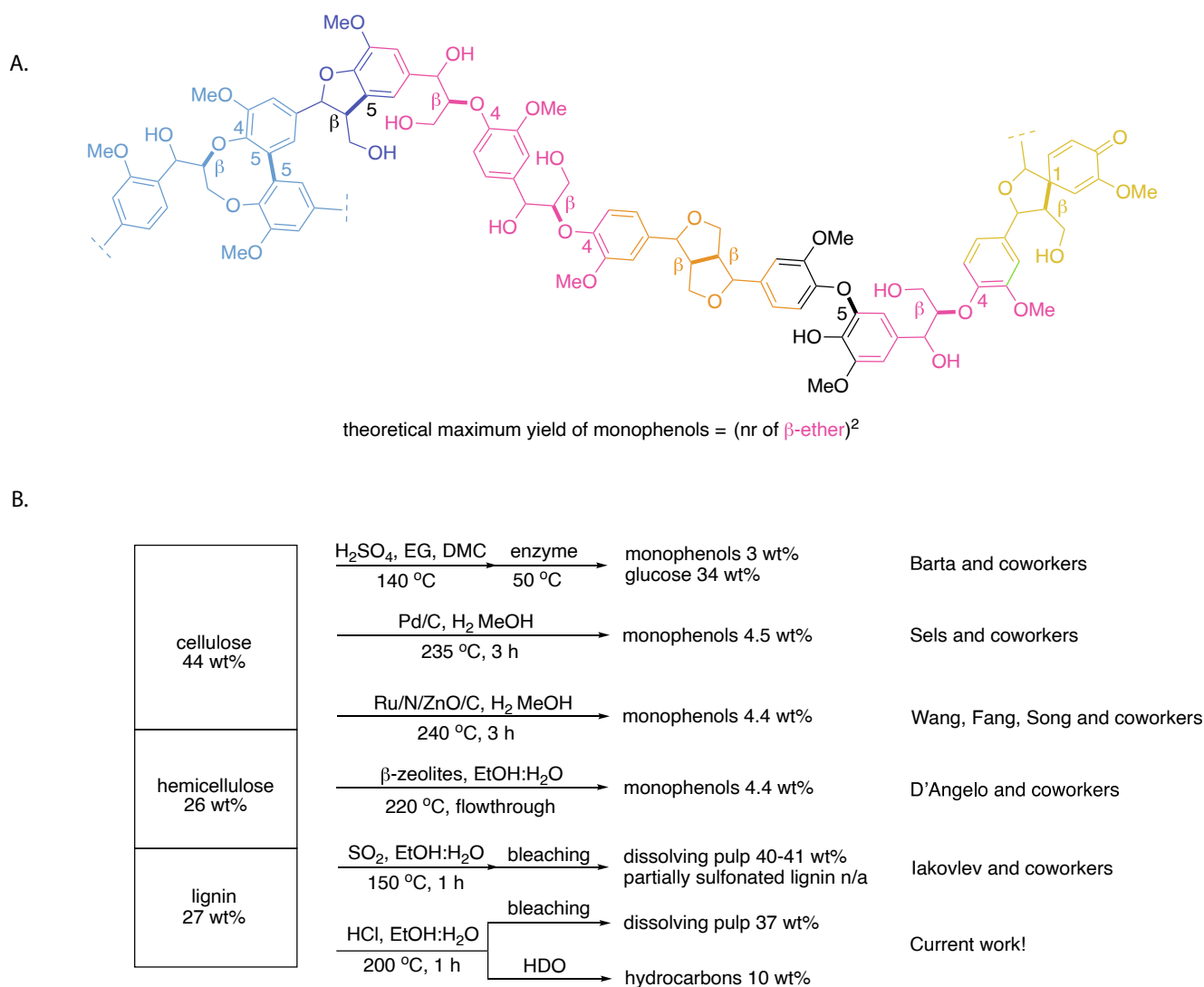

Fig. 1. A. Representation of softwood lignin; β-ether bonds in green are cleaved during depolymerization; theoretical maximum yield of monophenols depend on number of β-ether bonds in the native lignin polymer; **B.** Representative composition of softwood (major polymers); state-of-the-art in fractionation of softwoods. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

approaches on hardwoods, very few have been performed on softwoods which contain around 35–40% β-ether bonds, leading to considerably lower 'theoretical maximum yields' (12–16%) [24]. De Santi et al. demonstrated that 9 wt% of monophenols could be produced by a mild diol-assisted fractionation (DAF) employing acidolysis of pine wood in the presence of ethylene glycol (Fig. 1b) [28]. It should be noted that 9 wt%, which is close to the 'theoretical maximum yield', corresponds to 3 wt% of the original biomass. The pulp was enzymatically converted to glucose [29]. Van Aelst et al. performed RCF of pine wood using Pd/C and hydrogen at 235 °C for 3 h and obtained 16.7 wt% yield of mainly propanol guaiacol (Fig. 1B) [29]. The β-ether content was not measured, but the high yield indicates that other linkages than β-O-4′ were cleaved. The pulp fraction was not in focus of that study [30]. Liu et al. also reported yields of 14–16% for RCF of softwoods using a Ru-based catalyst running the reactions at 240 °C under hydrogen pressure for 4 h (Fig. 1B) [30]. The carbohydrate rich pulp was enzymatically degraded to glucose [31]. Kramarenko et al. performed a lignin-first approach in flow using β-zeolites and could obtain 10 wt% yield of monophenolic compounds running the reactions at 220 °C (Fig. 1B) [31]. Pan et al. pioneered in utilizing beetle killed wood and developed an organosolv fractionation to yield a pulp that was transformed to glucose (Fig. 1B) [32,33]. The lignin fraction was not a focus in these studies. Despite these advances, there are no reports on softwood lignin valorization to obtain beyond 5 wt% yield of monomers from biomass; only low value applications of pulp have been considered.

Organosolv pulping through acidolysis has been employed to fractionate wood to obtain dissolving grade pulp that can be used to produce textiles. However, either hardwoods or unbleached kraft pulps were used [34]. One notable exception is the sulfur dioxide ethanol water (SEW) fractionation that has successfully been applied to softwoods to produce dissolving grade pulp (Fig. 1B) [35]. However, the lignin generated is partially sulfonated which limits its applications, and no attempts to valorize this fraction was reported. To produce textile fibers have advantages from a life cycle perspective as compared to fermenting to glucose due to benefits related to carbon storage [36].

Due to the emerging environmental awareness on the impact of textile industry, there is a growing urgency to develop green and sustainable cellulose-based fibers. The use of secondary raw materials [37–39] and environmental friendly spinning processes [40,41], are being evaluated as strategies to reduce the dependency on cotton and to lower the environmental impacts of textile production. Lyocell fibers are considered as the most environmentally friendly fibers on the market [42], due to the cellulosic origin and the environmentally friendly process.

Recently, our group developed a value chain from hybrid poplar, a hardwood, to give dissolving grade pulp and a biofuel. However, the pulp was not regenerated and the lignin was upgraded using a non-conventional technology, employing formic acid as hydrogen donor without carrier liquid [43,44]. In addition, the land-use and climate change impact categories were not beneficial as marginal land was used for poplar plantations.

In order to realize a future biorefinery, both the carbohydrate and lignin are required to be valorized sustainably into high value products. Furthermore, industrially relevant set-ups and process windows should be used. It is important to develop methodology that is compatible with existing infrastructure to facilitate implementation. As mentioned above, only one study has reported upgrading of lignocellulose to both high value pulp from the cellulose-part and biofuel from lignin-part, however using unconventional hydrotreatment. Hydroprocessing of lignin is not trivial using industrially relevant process conditions. Important to note, only three studies have reported successful hydrodeoxygenation (HDO) of real lignin [45–47]. in other studies, mixtures of aromatic and phenolic compounds are obtained together with coke [48,49] –unless model compounds of monophenols have been used– where full deoxygenation has been reached [50–52]. Challenges with hydrotreatment of lignin, especially considering hydroprocessing using

a catalyst bed in current refinery set-ups, are associated with: polymeric nature of substrate; presence of oxygen, especially phenols that lead to coking; low solubility of lignin in hydrocarbon-based carrier liquids to facilitate substrate-catalyst interactions and thus control of reactivity. The solubility of the lignin in a hydrocarbon carrier is crucial to regulate the oxygen content of the feed and thus to control the exothermic reactions resulting from the HDO. Uncontrolled exothermic reactions lead to undesired coke formation [46]. Hence, processing of lignin in a current refinery set-up is only possible if the solubility issue is solved.

We hypothesized, and eventually demonstrated a holistic fractionation of beetle infected spruce, a softwood, to generate pulp that can be converted to textiles and hydrocarbons (Fig. 1B). To increase the sustainability and mitigate negative impact of land-use and climate change, beetle infected spruce was chosen as substrate. This raw material is considered a waste, as it is not accepted by pulp mills, and is incinerated to prevent spreading of disease [8,9]. In this work, beetle infected spruce was valorized to give dissolving grade pulp that was successfully spun to Lyocell fibers [53]. The lignin fraction was upgraded to biofuel using bio-derived furfural as a 'solubility-enhancing-agent' to promote dissolution in conventional carrier liquids. The sustainability of the value chain was evaluated by life cycle assessment (LCA). Our developed value chain is a major breakthrough in the valorization of softwoods and will inspire to use waste streams to generate value added products from both the carbohydrates and lignin fractions.

2. Experimental section

2.1. Materials and methods

All chemicals were purchased from Fischer chemicals, CCS Healthcare AB Sweden, Sigma-Aldrich, Honeywell, and VWR chemicals, and used as received. The biomass feedstock -beetle infected spruce (*Picea abies*)- was obtained from Mellanskog and was used for pulping. Gas chromatography coupled to mass spectroscopy (GC–MS) and GC with a flame ionization detector (GC-FID) analyses were conducted by a QP2020 system (SHIMADZU, Japan) equipped with two parallel HP-5MS columns (30 m × 0.25 mm × 0.25 μm). Gel permeation chromatography (GPC) analysis was performed on a Prominence-i, LC-2030C system (SHIMADZU, Japan) equipped with a UV detector a 280 nm. The IKA T-25 digital ULTRA-TURRAX® Homogenizer was used for the pulp disintegration.

2.2. General fast fractionation procedure

Beetle infected spruce (50 g, mix size < 5 cm) was loaded into stainless steel reactor (volume capacity 750 mL). The solvent mixture, 65% EtOH in water (400 mL), was added followed by HCl$_{aq}$ (0.35 M, 6.25 mL). The fast fractionation was performed at temperatures and reactions times specified in Fig. 2. After completion, the reaction was cooled down to room temperature, solid residue was filtrated and washed with water (200 mL) and EtOAc (200 mL). The water phase was extracted with EtOAc (2 × 100 mL). The combined organic phase was dried in *vacuo* to yield lignin oil, *vide infra*. The solid residue dispersed in 1 L of water and disintegrated (17.0 × 1000 rpm for 10 min) to give a homogeneous pulp. The solvent was drained and the pulp was oven dried at 60 °C for 12 h to yield a brown pulp. The pulp was bleached using NaClO$_2$ according to the procedure described in Supplementary text, Supplementary Materials.

2.3. Hydrodeoxygenation of lignin oil

Lignin oil obtained from fast fractionation (0.1 g) was placed in an autoclave (volume capacity 25 mL) together with furfural (0.5 g) and pentane (2 mL, 1.25 g), giving a lignin/furfural/pentane ratio of 1: 5: 12.5. Commercially available NiMo catalyst (50 mg) was added. The HDO reaction was performed at 360 °C (sand bath), the pressure of 25
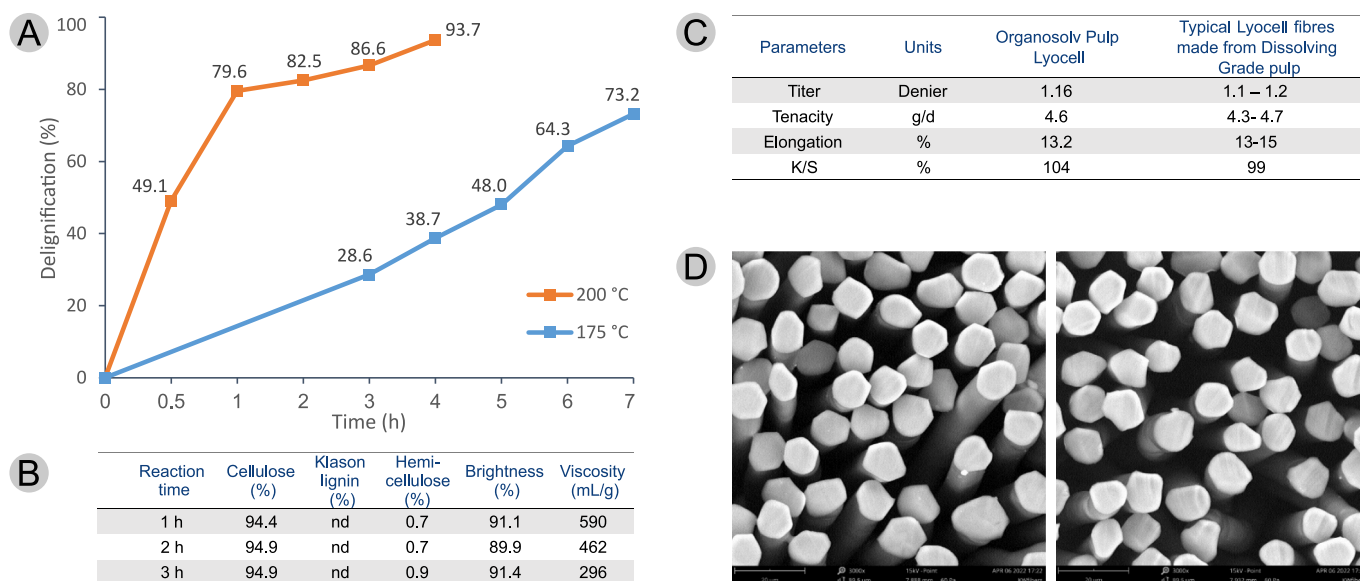
Fig. 2. Fast fractionation of beetle infected spruce. A. Degree of delignification over time of fast fractionation at pulping condition 200 °C (orange line) and 175 °C (blue line); **B.** Obtained dissolving grade pulp properties. nd = not detected; **C.** Quality of spun fiber compared to commercial sample; **D.** SEM images of fibers from fast fractionation of beetle infected spruce (both). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

B.

Reaction time	Cellulose (%)	Klason lignin (%)	Hemi-cellulose (%)	Brightness (%)	Viscosity (mL/g)
1 h	94.4	nd	0.7	91.1	590
2 h	94.9	nd	0.7	89.9	462
3 h	94.9	nd	0.9	91.4	296

C.

Parameters	Units	Organosolv Pulp Lyocell	Typical Lyocell fibres made from Dissolving Grade pulp
Titer	Denier	1.16	1.1 – 1.2
Tenacity	g/d	4.6	4.3- 4.7
Elongation	%	13.2	13-15
K/S	%	104	99

bar of hydrogen gas was applied at the beginning of the reaction. After 24 h, the reactor was cooled down and the pressure was released. The reaction mixture was collected and dried over anhydrous Na_2SO_4. The mixture was filtered to give a yellow oil. The generated oil was analyzed by 2D-GC and Simulated distillation according to the procedures described in Supplementary text, Supplementary Materials.

2.4. Life cycle assessment

Potential environmental impacts related to the conversion of beetle infected spruce into a regenerated cellulose, solid wood-based fuel and hydrocarbons in biofuel range were evaluated by a LCA where the four-phases framework standardized by ISO 14040 and ISO 14044 has been followed (1. Goal and scope definition; 2. Inventory analysis; 3. Impact assessment; 4. Interpretation). Details of the applied methodology are given in Supplementary text, Supplementary Materials.

3. Results and discussion

3.1. Fast fractionation of beetle infected spruce

3.1.1. Characterization of feedstock

The beetle infected spruce wood was obtained from Mellanskog, a forestry owner in Mid-Sweden and analyzed by different methods to determine the composition (Table 1, Supplementary Material, Table S1 for complete analysis): carbohydrates (62 wt%): hexoses (55 wt%): glucose 42.6 wt%, mannose 11.5 wt%, arabinose 0.9 wt%; pentoses (7

Table 1
Composition of beetle infected spruce.

Composition	wt%
Klason lignin, %	26.9
Glucose, %	42.6
Xylose, %	5.2
Mannose, %	11.5
Galactose, %	1.8
Arabinose, %	0.9
Ash, %	2.6
Extractives, %	8.0

wt%): xylose 5.2 wt%, galactose 1.8 wt%; lignin content 26.9 wt%; extractives 8 wt%; and ash 2.6%. β-O-4 content was determined by thioacidolysis to 36.9%.

3.1.2. Optimization of fast fractionation for textile fiber production

It is clear from state-of-the-art that focus to generate monophenols from cleavage of β-ether linkages would not be a sustainable fractionation approach for softwoods where<5 wt% overall yields from the biomass are anticipated. Carbohydrate valorization to ethanol *via* enzymatic hydrolysis gives yields below 25 wt%, resulting in overall mass balances below 30 wt% taking lignin valorization to monophenols into account. Thus, valorization to dissolving pulp has advantages: especially if the lignin fraction can be valorized beyond the 'theoretical maximum yield' of monophenols resulting from β-ether cleavage.

The fractionation experiments were performed on a 50 g scale of wood using a slightly modified literature procedure, with HCl as catalyst [54,55]. After the reaction was completed, the pulp was filtered off, ethanol was evaporated and lignin fraction extracted with ethyl acetate. Employing similar reaction conditions as Pan et al. used for beetle killed pine (170 °C, 65% EtOH, 1 h), a non-sufficient delignification was observed [32]. Even after 3 h, <30% delignification had occurred. To reach a practical delignification, 7 h were required, where 73.2% delignification was reached (Fig. 2A). However, the generated pulp was brittle, a characteristic of low viscosity due to degradation of the fibrils. We argued that a higher temperature would be beneficial. At 200 °C, the delignification was rapid and reached 79.6% within 1 h. After this, the kinetics of the delignification changed dramatically and after 4 h 93.7% delignification was reached. The viscosity of the generated pulp decreased from desired 590 mL/g observed after 1 h's reaction time to below 300 mL/g, when the reaction time was prolonged to 3 h and this is below the specifications (viscosity ≥ 450 mL/g) of dissolving grade pulp (Fig. 2B). Thus, optimized fractionation of beetle infected spruce was: performing the reaction at 200 °C for 1 h using HCl as catalyst (Fig. 2, A and B). A short reaction time is beneficial for the lignin fraction to prevent undesired condensation reactions, *vide infra*. The generated pulp was bleached and purified to meet the specifications of dissolving grade pulp (Fig. 2B, viscosity ≥ 450 mL/; brightness > 90%). To verify its applicability for regeneration, the pulp was successfully dissolved and spun using the Lyocell process, to obtain textile fibers of commercial

quality with respect to titer (1.16 Denier), tenacity (4.6 g/d) and elongation (13.2%) (Fig. 2, C and D). Thus, the fibers can substitute other regenerated fibers and also cotton in certain applications such as clothing.

3.1.3. Lignin-first approaches to yield monophenols

The organic fraction from fast fractionation was evaporated to give a lignin residue in 21.2 wt% yield. The monophenolics were directly isolated by dissolution in dichloromethane and quantified to 9.0 wt% yield (Fig. 3, A and B). This corresponds to 66% of the 'theoretical maximum yield' [21], which is a surprisingly high yield as no active stabilization technique, had been employed [20–22,24,56]. Vanillin and even monophenols with conserved propyl side chains were observed in the GC spectrum (Fig. 3A). Analysis of the lignin oil, by GPC, showed oligomers and polymers below Mw 6770 (Supplementary Material, Fig. S9) [57–59]. To increase the yield of monophenolic compounds, RCF in the presence of Pd/C was employed (Fig. 3B). Without any additional reductant, the yield of monophenolics did not increase [25,27,60]. When 10 bar hydrogen pressure was employed, slightly higher yields of monophenols were observed (13%), corresponding to 95% of 'theoretical maximum yield' (Fig. 3B) [61]. The most striking difference was the product distribution (Fig. 3A): Pd/C without additional reductant gave isoeugenols as major products; in presence of reductant aryl propane and ethane were the major products. In addition, the oligomeric fraction from RCF consisted of smaller oligomers (Mw < 2550) as compared to the lignin from fast fractionation (Mw < 6770) (Supplementary Material, Fig. S9). Thus, the product distribution can be steered by applying a stabilizing catalyst: however, the yield of monophenolic compounds does not motivate RCF [18,62]. Especially as upgrading of the pulp would require tedious purification from catalyst

or specialized equipment [43,60,63]. Another disadvantage of RCF –that has not yet been addressed by the community– is the ash content where the 2.6 wt% ashes present in the feedstock would contaminate any precious metal leading to poisoning of the catalyst and would not be economically or environmentally sustainable. The developed fast fractionation method without transition metals is sufficient and circumvents this. Nevertheless, 10–15% yield of lignin corresponds to < 5 wt% of the initial wood. Thus, another approach is required for softwoods to increase the value of the lignin fraction: while not sacrificing the valuable pulp.

3.1.4. Valorization of lignin fraction to biofuel by hydrodeoxygenation

We sought to convert the whole lignin fraction to a biofuel by hydrotreatment. As described in the introduction, one of the main challenges with hydrotreatment of lignin is its poor solubility in carrier liquids [46]. We argued that it would be worthwhile to use a bio-based 'solubility-enhancing-agent' to dissolve lignin in a hydrocarbon-based carrier liquid to facilitate hydrotreatment of the lignin. After initial screening, we found that furfural, generated from hemicellulose, readily dissolved the lignin in hydrocarbon-based carrier liquids. A commercial NiMo catalyst was used and the HDO was performed at 360 °C and 25 bar hydrogen gas (at RT) for 24 h in a batch autoclave reactor using a mixture of pentanes as a model carrier liquid [64,65]. Both lignin and furfural were fully deoxygenated to yield hydrocarbons. Furfural gave *n*-pentane, easily distinguished from the products obtained from lignin. The deoxygenated compounds from the hydrotreated oil were identified by 2D GC, and consisted of fuel components: Alkanes C_8-C_{18} in 12.1%, monocycloalkanes C_8-C_{12} in 18.9%, monoaromatics C_6-C_{12} in 20.8%, bicycloalkanes C_8-C_{13} in 4.8%, diaromatics C_{10}-C_{18} in 28.1% (Fig. 3C). The heavy fraction consisted of tricycloalkanes and polyaromatics in
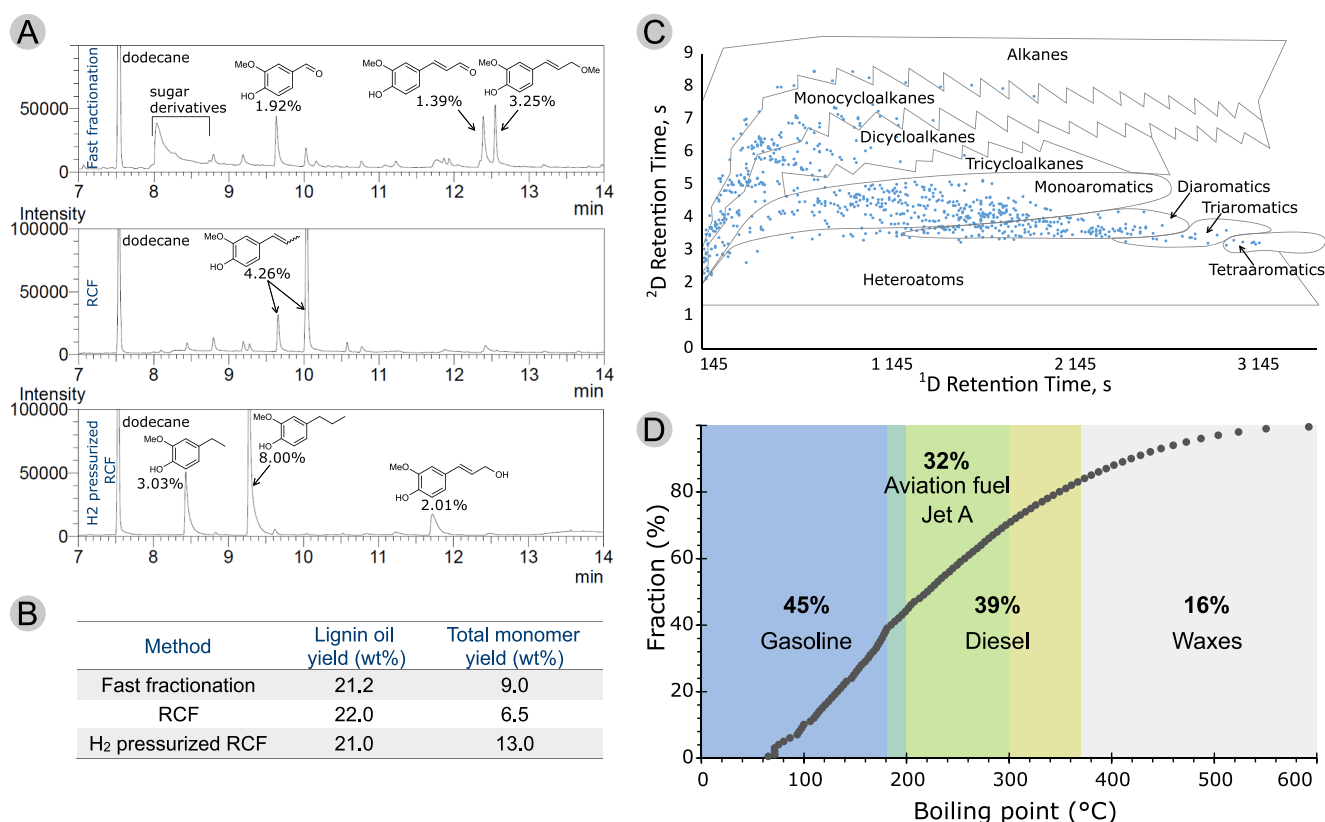

Fig. 3. Lignin fraction from fast fractionation A. GC spectra of monophenolic compounds from: top) fast fractionation; middle) reductive catalytic fractionation without hydrogen; bottom) reductive catalytic fractionation with hydrogen atmosphere; **B.** yield of lignin and monophenols from fast fractionation, RCF and RCF with hydrogen; **C.** GCxGC MS chromatogram of hydrotreated oil; **D.** Simulated distillation curve for the hydrotreated lignin oil (without pentanes), gasoline fraction (<200 °C) in blue, diesel fraction (200–370 °C) in yellow, aviation (Jet A) fraction(180–300 °C) in green, and waxes (>370 °C) in grey colour**.** (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

only 11%.

Simulated distillation (SIMDIST) of bio-oil after pentane deduction showed that the majority of products were found in the transportation fuels boiling point range (84%) comprising: 45% in gasoline range (b.p. < 200 °C) and 39% in diesel range (200–370 °C) (Fig. 3D) [66,67]. In addition, 32% of the products were in the boiling point range between 180 and 300 °C, that meets the requirements for aviation fuel, such as kerosene. Thus, the generated biofuel could meet a fluctuating demand of gasoline, kerosene and diesel. Hydrocarbons with boiling point above 370 °C constituted only 16% of the mixture and can be used as lubricants [67]. This fraction could be (hydro)cracked using more dedicated catalysts to obtain additional fuels if desired [68]. The aromatics content of the hydrotreated lignin fraction was > 50% resulting in high density and if used as aviation fuel alone it would have too high smoke point. Nonetheless, it would be an excellent blending stock with the prevailing biomass-derived aliphatic hydrocarbon products, including hydrocarbons from upgrading of *e.g.* triglycerides or from the alcohol-to-jet (ATJ) process. Due to the complete deoxygenation and high carbon retention in the liquid product phase, the energy content of the product, as characterized by its HHV is estimated to 45 MJ/kg, (Supplementary text, Supplementary Material).

3.2. Determination of overall mass balances

The mass balance of beetle infected spruce was calculated by combining the results obtained from fast fractionation at 200 °C for 1 h (Fig. 4). After the reaction, carbohydrate-rich pulp, which contained 89.1% of glucan, was separated as solid residue in 40.3 wt% from the starting biomass. The remaining lignin (5.5 wt%) and hemicellulose (2.6 wt%) were removed *via* bleaching to yield dissolving grade cellulose pulp in 36.6 wt%. It should be noted that this corresponds to 86% yield. Lignin oil obtained from fast fractionation contains monophenolic compounds up to 9.0 wt%, corresponding to 2.4 wt% of the initial biomass that could be recovered by distillation. To maximize lignin value, the whole lignin oil fraction was transformed by hydrotreatment to biofuel in 9.9 wt% yield from biomass. This corresponds to 60% of the theoretical maximum carbon yield considering inevitable losses of water and methane throughout the HDO. However, the methane is reformed to hydrogen in the refinery, and this makes up a substantial amount of the hydrogen used in the HDO (Supplementary Material, Table S9). During

pulping, hemicellulose and some part of cellulose were hydrolyzed and gave a mixture of pentose and hexose derivatives in 13.9 wt% that were isolated and characterized from the aqueous phase and could be used as a make-up solvent for losses in the fractionation step (Fig. 4; Supplementary Material, Table S9). Overall, this process yields dissolving grade pulp in 36.6 wt%, a mixture of C5 and C6 sugar 13.9 wt%, and biofuel up to 9.9 wt%.

3.3. Life cycle assessment of value chain

To determine the environmental sustainability of the proposed value chain, a comparative LCA was performed to relate the obtained regenerated cellulose to cotton production. Cotton was chosen because it is the most used fiber and has a similar function and chemical composition as regenerated cellulose (See Supplementary text, Supplementary Materials for further discussion) [69,70]. A cradle-to-gate LCA of the value chain including harvest, and transport of beetle infected spruce from forest, pulping and bleaching biomass to dissolving grade of cellulose and regeneration of the fiber through viscose method has been performed. The choice of cradle-to-gate system boundaries was made because the subsequent steps are highly dependent on the specific application and their treatment would require to make significant assumptions (further discussion of this aspect is given in Supplementary text, Supplementary Materials). Furthermore, as the generated fiber meets the specifications of Lyocell fiber, there should not be any deviations in use-phase or end of life from a commercial fiber. Secondary data for viscose was used as data for the Lyocell process is not available. 1 kg of viscose fiber was chosen as functional unit. By-products such as bark and reject fiber as well as the biofuel generated through hydrotreatment were treated by substitution approach, crediting incineration to produce power and heat and fossil fuel for transportation respectively [71]. Impact assessment was performed with a focus on the environmental footprint categories: Climate change, *i.e.* Global warming potential, Land use, Water use and Resource use, fossil and mineral (Supplementary Material, Table S8). The full set of impact categories of the Environmental Footprint 3.0 (EF) method was also studied to mitigate burden shifting (Supplementary Material, Table S8) [72]. To increase robustness of the assessments, the ReCiPe 2016 Hierarchist method was used as a part of the sensitivity analysis, where similar results were obtained (Supplementary Material, Table S14) [73]. These impact categories were quantified for viscose fibers from the beetle infected spruce value chain and compared to cotton fibers at ginning stage produced from current farming practices and production technology [74]. As end of life can be considered similar for both systems, this phase was not included in the assessment. A detailed description of the LCA goal, scope, calculations, and assumptions can be found in Supplementary text, Supplementary Materials.

To assess the fiber production, inventory analysis to produce 1 kg of viscose fiber was collected using primary data for determining yields of pulping and HDO and secondary data for the other unit operations for both generation of viscose fiber from beetle infected spruce as well as production of cotton fiber (Fig. 4). To study the energy demand of the pulping, a 6:1 solvent to wood ratio was used (state-of-the-art) (Supplementary Material, Table S9) [75]. Secondary data was used for the inventory analysis of cotton production.

Four of the five environmental footprint impact categories gave lower environmental impact for the viscose from beetle infected spruce compared to cotton production (Fig. 5A). The only impact category that gave a higher impact was the resource use, minerals and metals. This is because of the consumption of zinc concentrate for the production of sulfur dioxide used in the viscose process (Fig. 5B). For climate change, land use and water use, the impact was significantly lower than cotton production and also in comparison to the value chain from poplar [25]. It should be noted that the starting material, namely biomass from beetle infected spruce, must be removed from the forest: so that the system analyzed does not create additional pressure on land use change.
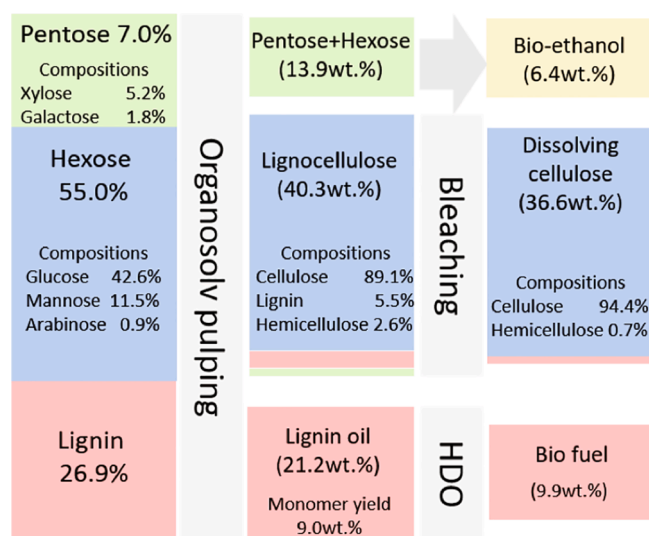

Fig. 4. Mass balance of the beetle infected spruce during fast fractionation. Numbers are reported as weight percent (wt%) relative to biomass. Optimized reaction condition: Beetle infected spruce (50 g), 0.36 M HCl$_{aq}$ (6.25 mL, 1 wt% to biomass), 65% EtOH (400 mL), 200 °C, 1 h (excluding 20 min to reach 200 °C from room temperature).
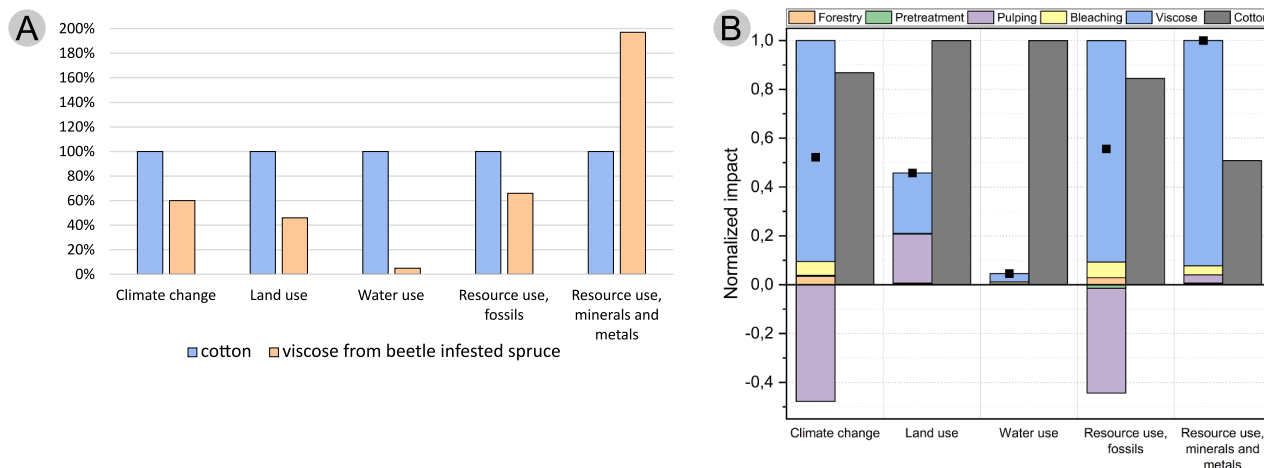
Fig. 5. A. Footprint categories of regenerated fiber from beetle infected spruce compared to cotton; **B.** Contribution of different process stages in the production of textile fiber from beetle infected spruce and cotton.

Similarly, biodiversity is not affected, however, this is difficult to assess as the indicators are not fully developed in LCA [76]. Most significant was water use, which is a known problem for cotton production. Here, the impact of viscose from beetle infected spruce was only 5% of the cotton fiber.

4. Discussion

To meet a forecasted 25% population-increase predicted to 2060, without de-forestation; with crude oil exhausted, appears overwhelming. Textiles are currently produced from cotton and fossil-derived synthetic materials. Cotton production alone, uses 34.5 million hectare of arable land worldwide to yield 26 million tons of cotton lint [77]. The industry consumes 77 million m^3 of blue water [78]. Furthermore, it has been predicted that cotton production cannot increase further due to restrictions in land use [79]. Regenerated cellulose from forestry can replace an important fraction of produced cotton [70]. However, current forestry is also a limited resource. Thus, using forestry residues –that today are considered wastes– for production of textile fibers and biofuels, could contribute to supply a future demand in a sustainable way. The high quality Lyocell fibers from beetle infected spruce developed in this work, after extraction of lignin oil is a significant advancement towards next generation of sustainable fibers.

Only in Europe, 100 M m^3 of wood is destroyed annually [15]. Thus, 42.3 million tons of wood could potentially be upgraded to benign fibers for textiles. With a 36 wt% yield (86% theoretical yield), 15 million tons of regenerated cellulose could be produced only in Europe and this exceeds the predicted demand from population growth during this century. This is equivalent to 20 million hectares of agricultural land that would be required for cotton production. In addition, 4 million tons of biofuel would be co-produced. Taking into account that this value chain does not compete with food production or forestry, there is potential to meet textile needs of future generation without negative impacts on climate change, land-use or water scarcity by employing the proposed value chain and this was also confirmed by LCA. It would be worthwhile to perform a cradle to grave LCA study in the future for a specific application using the produced fiber.

The value chain developed in this study thus aligns with several of the 17 UN Sustainability Development Goals, such as: 2, Zero hunger; 3, Good health and well-being; 6, Clean water and sanitation; 7, Affordable and clean energy; 8, Decent work and economic growth; 9, Industry, innovation and infrastructure; 12, Responsible consumption and production; 13, Climate action; 14, Life below water; 15, Life on land. The value chain is in accordance to both the European Green Deal and American Inflation Reduction Act that target environmental sustainability including climate change, land use, water scarcity and resource depletion.

5. Conclusions

We successfully developed a novel value chain from beetle infected spruce that currently is burnt to a low value. Valorization of the raw material was performed by converting the raw material to a pulp that could be further transformed into high value Lyocell textile fibers and hydrocarbons in gasoline-aviation-diesel range. Key developments comprise: fractionation of a softwood to obtain both a dissolving grade pulp that was spun to textile fibers; high monophenol yield without catalyst. Lignin was upgraded to biofuels by employing a bio-based 'solubility-enhancing-agent' to enable hydrotreatment. By this strategy, 10% of the original biomass was converted to biofuels, corresponding to 60% carbon yield. The environmental impact of the developed value chain was evaluated by LCA. Of the five most important environmental footprint categories, the proposed value chain performed better than cotton fiber production in four. From an environmental perspective, this is a promising way to mitigate challenges with a predicted increase in population. This study makes progress in the research field by demonstrating holistic valorization of both carbohydrate and lignin fractions in a softwood; positively assessed by LCA. We hope that this study inspires researchers to further development of valorization of residues that today are burnt to meet current and future demands.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Joseph Samec and Alessandro Manzaro reports a relationship with Stockholm University that includes: board membership. Joseph Samec has patent pending to assignee. JS is also co-founder of RenFuel; Alessandro Manzardo is co-founder of Spin-life.

Data availability

Data will be made available on request.

Acknowledgements

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2023.144179.

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