



Techno-economic analysis of thermal deoxygenation based biorefineries for the coproduction of fuels and chemicals

Sampath Gunukula^{a,b}, Sharon J.W. Klein^{b,c}, Hemant P. Pendse^{a,b}, William J. DeSisto^{a,b}, M. Clayton Wheeler^{a,b,*}

^a University of Maine Chemical and Biomedical Engineering, Orono, ME 04469-5737, USA

^b Forest Bioproducts Research Institute, Orono, ME 04469-5737, USA

^c School of Economics, Orono, ME 04469-5737, USA

HIGHLIGHTS

- The economic viability of thermal deoxygenation pathway was assessed.
- The economic benefit of brownfield biorefinery site was analyzed.
- The profitability of activated carbon production from biochar was determined.
- The impact of co-production of chemicals and fuels was assessed.

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ABSTRACT

The economic impact of coproduction of renewable fuels with platform chemicals is determined by assessing thermal deoxygenation (TDO) pathway based biorefineries with two distinct product suites: (1) fuel and furfural (a platform chemical) and (2) fuel and levulinic acid (a platform chemical). Greenfield and brownfield plant site scenarios are considered. Additionally, the economic viability of producing activated carbon from biochar, rather than burning it to produce energy, is assessed. The biorefinery in which wood is converted using the TDO pathway to furfural, renewable fuel, and biochar is found to be a product-driven biorefinery. The production of renewable fuel along with levulinic acid from recycled cardboard via the TDO pathway is determined to be an energy-driven biorefinery. The minimum selling price (MSP) of TDO oil produced in product-driven and energy-driven greenfield biorefineries are estimated at US\$17 and US\$18 per GJ or US\$2.78 and US\$2.92 per gallon of gasoline equivalent (GGE), respectively. It is found that capital investment for the product-driven and energy-driven biorefineries can be reduced by 23–27% through integrating the TDO process into an existing pulp and paper facility (brownfield site), provided the infrastructure and assets of pulp and paper mill are well maintained. It has been learned that the synergy arising through co-production of chemicals with fuels and vice versa can facilitate the sustainable production of both renewable fuels as well as platform chemicals. Moreover, there is a need for the development of new less energy intensive separation processes for purifying bio-products.

1. Introduction

The transportation fuels and chemicals used to produce consumer products are predominantly made from natural gas and crude oil [1]. The extraction of these non-renewable feedstocks emits greenhouse gases (GHG) that cause global warming. To reduce GHG emissions of transportation fuel and chemical industry, as well as to reduce dependence on volatile petroleum feedstock markets, technologies have been proposed to produce fuels and chemicals from a renewable feedstock [1]. The fast pyrolysis and catalytic fast pyrolysis pathways have been

extensively investigated to synthesize transportation fuels from renewable lignocellulosic feedstocks [2,3]. The US Department of Energy introduced the concept of bio-based building block or platform chemicals from which a wide range of secondary commodity chemicals can be made to accelerate the growth of biorenewable chemical industry [1,4]. However, achieving economic viability of both renewable fuel and platform chemical production remains a challenge [5,6]. Such a challenge can be overcome by developing biorefineries in which new technologies can be used to convert biomass to both transportation fuels and platform chemicals [7]. The strategy of producing both

* Corresponding author at: University of Maine Chemical and Biomedical Engineering, Orono, ME 04469-5737, USA.
E-mail address: mcwheeler@maine.edu (M.C. Wheeler).

platform chemicals and fuels in a biorefinery has not been assessed yet and thus there is a need to determine the economic advantage of this strategy.

A biorefinery is a facility in which renewable feedstock is sustainably converted to a wide spectrum of marketable bio-based products [8,9]. Biorefineries can be classified into product-driven and energy-driven categories [9]. In the product-driven biorefinery, relevant to the chemical and fuel industry, the major product (in terms of revenue generated) is high market volume (platform) chemicals and the co-product is energy, such as biofuel and electricity [6]. In the energy-driven biorefinery, the renewable feedstock is used to produce energy as a major product and high-value-added platform, secondary-commodity, and specialty chemicals are minor products [9]. Determining how co-production of chemicals with biofuels, and vice versa, can influence the overcoming economic barriers of biomass conversion processes is necessary to guide future research for the development of economically viable biorefineries [10].

Biorefineries can be constructed on a new plant site (greenfield) or through repurposing existing industrial facilities such as pulp and paper mills (brownfield) [11]. The financial and market risks to biorefinery investments and the requirement of a high amount of capital for constructing greenfield biorefineries are delaying the commercialization of biorefinery projects [11,12]. It is expected that the capital investments for a new biorefinery can be reduced through constructing brownfield biorefineries [11]. However, the percentage of capital cost savings with brownfield biorefineries built around thermochemical pathways for the production of renewable fuels has not been assessed yet [13–15]. Thus, there is a need for assessing capital investment for integrating the biorefineries built around thermochemical pathways into the selected industrial facility. Furthermore, this assessed capital cost must be compared against the capital investment for the greenfield biorefinery to determine the repurpose potential of the brownfield biorefinery in terms of percentage of capital cost savings.

Biochar is a common by-product of all lignocellulosic biorefineries [4,16]. Generally, biochar is slated to burn for meeting energy needs of the biorefinery, and the excess energy can be sold to an electricity grid [16]. Recently, it has shown that biochar can be upgraded to a range of high value-added biomaterials [17–20]. One such biomaterial is activated carbon [16]. The biochar can be upgraded to activated carbon with a high surface area using either physical or chemical activation methods [17,21]. The impact of the co-production of activated carbon from biochar on the profitability of biorefinery investments has not been determined yet [13,22]. The market price of activated carbon is higher than that of biochar and that the production of activated carbon can generate higher revenues [17,21]. However, there is a significant loss of biochar during the upgrading process [17,21]. Moreover, there are capital and operating costs associated with the production of activated carbon [17]. Thus, the economic feasibility of upgrading excess biochar to activated carbon in biorefineries must be determined.

Fast pyrolysis and catalytic fast pyrolysis to make renewable fuels fall under the category of energy-driven biorefineries [3,4]. The higher capital costs associated with the use of expensive catalysts, rapid deactivation of catalysts, use of hydrogen, and the lack of co-product revenue are limiting barriers for achieving commercialization of these pathways to make renewable fuels [3,4]. Such barriers may be overcome by using acid hydrolysis and dehydration (AHDH) combined with thermal deoxygenation (TDO) [23–29]. The AHDH process involves the conversion of C₆ sugars to organic acids [23–26]. In the TDO process, the organic acids are first neutralized either with calcium or magnesium salts [27–29]. These neutralized organic salts later decompose to a renewable fuel with a low level of oxygen content at a high temperature [27–29]. The capital investment to produce renewable fuels via the TDO pathway could be lower than that of fast pyrolysis and catalytic pyrolysis pathways because the production of TDO oil does not require an expensive catalyst and hydrogen production. However, the separation and purification of organic acids from the dilute product stream

originating from the AHDH reactor could demand high capital and operating costs. Thus, performing a detailed economic and modeling analysis is necessary to assess the commercial feasibility of the production of renewable fuels via the integration of the AHDH and TDO processes.

In the current study, we performed the economic and modeling analysis of biorefineries built around the integration of AHDH and TDO processes for the conversion of wood and cardboard to assess economic viability of integration of the AHDH and TDO processes. The economic impacts of co-production of biofuels with platform chemicals and vice versa were investigated by analyzing the results of economic performance of the biorefineries. The impact of the production of chemicals with different market demands on the economics of the biorefinery, and the economic viability of upgrading excess biochar to activated carbon were assessed. Finally, the potential capital cost savings by repurposing an existing well maintained industrial facility to make renewable fuels and chemicals via the TDO pathway were quantified.

2. Process models of biorefineries built around the TDO pathway

The integration of AHDH and TDO processes (from now on simply the TDO pathway) offers flexibility in the use of feedstock that enables us to construct biorefineries with distinct product suites of chemicals and fuels. The lignocellulosic feedstock (e.g., wood, corn stover) is heated in dilute sulfuric acid to produce levulinic acid (LA), formic acid (FA), furfural (FUR), and biochar. FUR (a platform chemical) can be sold at a high market price. A fraction of biochar can be upgraded to activated carbon and the remaining fraction can be burned to meet the energy requirement of the biorefinery. The LA and FA are neutralized with calcium hydroxide to form calcium levulinate and calcium formate, respectively. The calcium salts of organic acids are decomposed to produce TDO oil. The other possibility is before neutralization LA and FA can be purified to sell small fractions of these purified products as chemicals. However, the optimal molar ratio of FA and LA to make high quality TDO oil with a low level of oxygen content is one and thus selling a fraction of FA as a chemical can lead to the production of low quality TDO oil [30]. A recent work [31] has shown that multiple solvent extraction columns and energy intensive distillation columns are required to extract LA from the mixture containing FUR, LA, FA, water, and H₂SO₄, which increases the process complexity as well as capital and operating costs. Thus, producing FUR and TDO oil in the biorefinery seems to be the best option among possible scenarios. Section 2.1 provides the detailed process description of the production of TDO oil and FUR.

Cellulose rich feedstock such as cardboard or starch based feedstocks such as corn and cane sugar can be used to produce LA and FA via AHDH. A small fraction of the LA (a potential platform chemical) can be sold as a co-product and this fraction is highly dependent on the market price of LA as well as its market demand. The future market demand and market growth of LA has not been defined as the technologies for the conversion of LA to fuels and chemicals are still at the development stage. The FA and the remaining fraction of LA are converted to TDO oil as described previously. The FA can be purified using multiple distillation columns to sell a fraction of the total FA produced as a chemical when cellulose-rich feedstock is used. However, it is not possible to achieve a high purity of FA without affecting separation yields and the quality of TDO oil [30–33]. Section 2.2 provides the relevant process differences when producing only TDO oil and LA.

2.1. Production of FUR and TDO oil

A simplified block flow diagram (BFD) was developed for the conversion of blended woody feedstock (cellulose content of 45%, hemicellulose content of 22.5%, lignin content of 22.5%, extractive content of 8%, and ash content of 2%) to FUR, TDO oil, and biochar via the TDO pathway (Fig. 1). Detailed process flow diagrams are included in the supplementary material.

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