



Research article

Techno-economic analysis of utilization of stillage from a cellulosic biorefinery



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ABSTRACT

Cellulosic biorefinery stillage contains waste water with dissolved unutilized fermentable sugars and, mainly, lignin of the biomass. Currently, the best use of this lignin is through direct combustion to supply industrial process energy; however, fast pyrolysis has recently attracted researcher's interest as it has the potential to convert lignin into bio-oil and bio-char. It is essential that these alternative routes be thoroughly analyzed for their techno-economic feasibilities and bottlenecks, which was the main objective of this study. Stillage from a cellulosic biorefinery of a 113.5 million liters per year (30 million gallons per year) butanol production facility was considered in this study. Experimental and modeling data for both fast pyrolysis and direct combustion systems were gathered from recent publications and used for analysis. Modeling software, SuperPro Designer, was used to develop process models and economic analyses of both systems. The estimated stillage processing cost (\$/1 butanol produced) of a direct combustion system and fast pyrolysis system were found to be 0.15 and 0.17, respectively, including byproducts credits. Plant size, anaerobic digester retention time, the moisture content of solid stillage, turbine and boiler efficiencies, and cost of waste treatment chemicals were the most sensitive input parameters. At the present state of these technologies, direct combustion is a more economically feasible option than fast pyrolysis; however, both could be of interest to commercial cellulosic biorefineries, interested in lignin utilization alternatives, due to the marginal difference in stillage utilization costs.

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1. Introduction

Cellulosic biorefinery utilizes lignocellulosic biomass, such as agricultural residues, forest residues and energy crops, to transform into biofuels, which can substitute gasoline and have environmental benefits over fossil fuels. Thus, globally, production of biofuels has been supported by several governments, including the U.S. government, which has mandated 16 billion gallons of biofuels be produced from lignocellulosic biomass by 2022 [1]. Cellulose (30–50 wt%), hemicellulose (8–50 wt%) and lignin (7–30 wt%) are major constituents of lignocellulosic biomass [2,3]. Variations in their respective compositions are due to biomass types, maturity, harvesting season, and climate [4]. Generally, hardwood consists of larger fractions of cellulose and lignin, while most agricultural residues and energy crops have average of these extreme compositions.

During the biofuel production process, a cellulosic biorefinery could take the lignocellulosic biomass as an input, which is then pretreated (e.g., commercial sulfuric acid pretreatment) to destroy the lignin barrier and transform most of the hemicellulose and some fraction of

cellulose, for instance, about 60–90 wt% and 3–10 wt%, respectively, into fermentable sugars [4,5]. Additionally, about 5 wt% of the lignin would be converted into acid soluble lignin during the sulfuric acid pretreatment process [4]. Generally, the rate of cellulose and hemicellulose degradation depends upon the type of pretreatment processes such as chemical, nonchemical and biological [6]. The remaining cellulose and hemicellulose fractions are further treated with an enzyme called cellulase, where most of the remaining cellulose and hemicellulose, i.e., about 90–96 wt%, transform into fermentable sugars [4,5]. Following this, the fermentable sugars are transformed into biofuels during successive microbial fermentation processes. Thus, in addition to lignin, a small fraction of the unused cellulose and hemicellulose also remains in the waste stream. A recent study [7] analyzed wheat straw solid stillage composition, and found about 79.0 wt% lignin, 8.3 wt% cellulose, and 3.6 wt% hemicellulose. A techno-economic study of a corn stover based ethanol plant, using sulfuric acid pretreatment, [4] reported that the cellulosic biorefinery waste streams contained 74.1 wt% of lignin followed by 7.5 wt% cellulose and 3.1 wt% hemicellulose based on the remaining total solid fraction of the biomass. The lignin fraction of the feedstock biomass was reported to be about 95% lignin [4]. These unutilized materials remain as the major solid fraction in the waste stream of biorefineries.

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Apart from the solid fraction of stillage, during the biofuel production process, about 15 gallons of process water is required for each gallon of ethanol produced [4]. Unutilized fermentable sugars during fermentation are major fraction that is dissolved in the wastewater and remained in the waste stream. A previous study [4] reported that about 22% of the fermentable sugars that were present before fermentation remain in the waste stream. Overall, a cellulosic biorefinery stillage contains about 87.2 wt% wastewater, 7.8 wt% process chemicals, 3.6 wt% biomass, mainly lignin, and 1.4 wt% fermentable sugars [4].

Stillage has pollution potential due to the presence of high-strength process chemicals and organic matters [8] requiring some treatment before discharging into a sewer system or watercourse. Economic benefits can be generated from the stillage by transforming the solids and dissolved fermentable sugars into useful forms of energy. Additionally, much of the wastewater can be recovered. However, a previous study [4] reported that about 55% of the total plant equipment cost was required for wastewater treatment through anaerobic followed by aerobic treatment and the solid utilization through direct combustion. Electricity and process steam were the main products from this stillage treatment process. To date, even established cellulosic industries are using direct combustion to recover energy from lignin. For example, only about 2% of the 50 million tons of lignin produced annually from the pulp and paper industry is used for commercial products, while the remaining lignin is used to produce process steam and electricity through direct combustion [7]. Thus, due to capital intensive stillage treatment process, conversion of lignin into more valuable products than electricity and process steam is essential for economic and sustainable biofuel production in the future.

With the emerging interest in commercial cellulosic biorefineries, concern about the efficient utilization of the lignin waste has increased rapidly. In general, lignin has a complex structure formed by a number of chemical groups such as aromatic rings, phenolic, aliphatic, alcohol and methoxy groups [7,9]. Currently, interest in fast pyrolysis of lignin is growing as it can directly produce bio-oil, which can be further upgraded into biofuels, such as gasoline, diesel, and jet fuel, and bioproducts, such as bio-asphalt, carbon fiber, and chemicals for industrial and medical applications [10,11]. In addition to bio-oil, bio-char produced during fast pyrolysis can be added to soil to increase the soil carbon content and maintain soil properties that are very sensitive to biomass removal from the field [12]. Further, a more valuable product than bio-oil needs to be produced from the lignin fraction. Previous studies [13,14] reported that the economic and environmental sustainability of a cellulosic biorefinery depends on utilization of the lignin fraction to produce valued-added chemicals, such as aromatic chemicals, phenols, benzene, toluene, xylene and vanillin. Previous study [13] also reported that catalytic lignin depolymerization using triflic acid had the potential to produce commercial aromatic chemicals. Additionally, different bond cleavage strategies for lignin depolymerization are discussed in a recent review [15]. Currently, Borregaard, an international company, commercially produces vanillin from lignin, which has a market value of 100–200 \$/kg and is in high demand by the perfume industry, European chocolate manufacturers and Japanese markets [14]. Further studies are needed to evaluate the economic feasibility of these options, particularly utilizing lignin from cellulosic biorefineries.

Apart from the solid fraction, while evaporation of the liquid fraction requires an extensive amount of energy [16], anaerobic digestion requires less energy and might be a good option for removal of the residual organic matter in the stillage by converting it to biogas [4,5]. The biogas can be further transformed into electricity. The wastewater from anaerobic digestion can be further passed through aerobic treatment to recover the water [4]. Thus, solid utilization through both direct combustion and fast pyrolysis, and anaerobic digestion of the liquid fraction of stillage followed by aerobic wastewater treatment were analyzed in this study. Two processes were analyzed: (1) a direct combustion system (DCS), which included both direct combustion of the solid fraction of stillage and a wastewater treatment process; and (2) a fast

pyrolysis system (FPS), which included fast pyrolysis of the solid fraction of stillage and an identical waste water treatment process.

While the DCS is an established technology, in the past 20 years, extensive efforts have been made to carry out fast pyrolysis of lignin at laboratory scale to establish the potential of this method for lignin processing and to compare procedures and products [7,9,17–19]. However, a detailed techno-economic analysis to assess the technical and economic feasibilities of establishing commercial scale fast pyrolysis using lignin from cellulosic biorefineries has not yet been done. With the development of commercial cellulosic biorefineries, there is an immediate need to explore the techno-economic feasibility of fast pyrolysis of lignin in the waste stream from bioprocessing. This may reduce the overall biofuel production cost due to the opportunity for commercial utilization of high value products, i.e., bio-oil and bio-char. Moreover, a techno-economic study is needed for comparison of direct combustion and fast pyrolysis of lignin. Although small scale studies have shown significant progress in the use of pyrolysis, the commercial feasibility of high value applications for lignin have not been fully quantified; thus, a detailed techno-economic analysis is needed to compare direct combustion and pyrolysis of lignin from the cellulosic biorefinery waste stream. Thus, the main goal of this study is to analyze the techno-economic feasibilities, and bottlenecks of both DCS and FPS. The outcomes of this work could provide commercial benefits for the cellulosic biorefinery and pulp and paper industries.

2. Methods

2.1. Modeling overview

The expected stillage from a cellulosic biorefinery with an annual butanol production capacity of 113.5 million liters per year (MLPY) (i.e., 30 million gallons per year (MGPY)) as proposed by Baral and Shah [20] was considered in this study. As discussed earlier, stillage mainly contains waste water, lignin, and unutilized sugars. Table 1 summarizes the chemical composition of the stillage [20]. The process models of the fast pyrolysis and direct combustion systems were developed using modeling software, SuperPro Designer [21]. The general overview of both systems with process description is summarized in

Table 1
Composition of stillage from ABE fermentation.

Component	Mass (kg/kilo-liter butanol produced)	Mass composition (wt%)
Acetic acid	4.43	0.02
Acetone	8.48	0.04
Ammonium sulfate	230.75	1.19
Ammonium acetate	137.38	0.71
Ash	296.25	1.52
Butanol	44.65	0.23
Butyric acid	2.49	0.01
Enzyme	37.93	0.20
Cellulose	578.31	2.98
Inoculum	1129.34	5.81
Ethanol	1.27	0.01
Furfural	41.36	0.21
Glucose	478.80	2.46
Hemicellulose	126.58	0.65
Hydroxymethylfurfural (HMF)	8.84	0.05
Lignin	848.47	4.37
Nutrient medium	211.24	1.09
Other solids	7.37	0.04
Protein soluble	183.31	0.94
Soluble extractive	636.25	3.27
Soluble lignin	173.91	0.89
Soluble solids	59.76	0.31
Water	13,761.61	70.81
Xylose	426.40	2.19
Total	19,435.19	100.00

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