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Sustainability assessment of cellulosic biorefinery stillage utilization methods using emergy analysis

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ABSTRACT

Energy can be recovered from stillage from cellulosic biorefineries in different ways, including direct combustion and fast pyrolysis. These different energy conversion routes require different level of inputs from natural resources, non-renewable resources, and economic services. Due to the high energetic and economic costs of stillage recovery methods, it is essential to perform a sustainability analysis of these different options before commercial deployment. Thus, the main objective of this study was to assess the relative sustainability and environmental impact of fast pyrolysis and direct combustion systems for the beneficial use of waste stillage using emergy analysis. The estimated emergy sustainability indices of direct combustion and fast pyrolysis were 0.09 and 0.07, respectively, where the renewable fraction of stillage was the most influential input parameter. Additionally, the net product transformity for direct combustion and fast pyrolysis were 7.06E+05 and 2.61E+05 seJ/J, respectively. Overall, a 23% higher emergy sustainability index for the direct combustion compared to the fast pyrolysis and a 63% lower overall product transformity for the fast pyrolysis compared to the direct combustion suggests that both systems, at the current state of the technology, offer differing advantages for stillage utilization depending upon the desired end products and uses.

1. Introduction

Several countries have policies that support the development of biofuels from lignocellulosic biomass, such as agricultural residues, forest residues, and energy crops. For instance, the U.S. government has mandated that 16 billion gallons/year of cellulosic biofuels, such as ethanol and butanol, be produced from lignocellulosic biomass by 2022 [1]. These cellulosic biofuels can be used as direct substitutes for gasoline and are claimed to have environmental benefits over fossil fuels [2,3]. The major constituents of lignocellulosic biomass are cellulose (30–50 wt%), hemicellulose (8–50 wt %) and lignin (7–30 wt%) [4,5]. During biochemical conversion of biomass into biofuels, most of the cellulose and hemicellulose are first transformed into fermentable sugars and then into biofuels; lignin, however, cannot be utilized by microbes and thus remains in the waste stream. This process requires about 15 L of process water per liter of ethanol produced [6], which is also a component of the waste stream. A recent study [7] found that about 79 wt% of lignin remains in the cellulosic biorefinery waste stream followed by 8.3 wt% cellulose and 3.6 wt% hemicellulose. Besides these solid fractions, the waste water contains unutilized fermentable sugars and process chemicals (Table 1). Both solid and liquid wastes of cellulosic biorefineries are collectively known as stillage. A recent techno-economic study of a commercial scale cellulosic ethanol production system with sulfuric acid pretreatment [6] reported that about 22.12% of fermentable sugars that were present before fermentation remain in the stillage. That study [6] also reported that about 43% of total plant cost is required for stillage utilization through direct combustion process. Thus, a low cost and low energy stillage recovery method is essential for economic and sustainable biofuel production in the future.

As discussed earlier, the stillage is a high-strength effluent due to process chemicals and organic matters, which have pollution potential [8,9]; thus, it can neither be sent to the sewer system nor be discharged into a water body or soil. Nevertheless, the stillage

Abbreviations: DCS, Direct combustion system; EIR, Emergy investment ratio; ELR, Environmental loading ratio; ESI, Emergy sustainability index; EYR, Emergy yield ratio; FPS, Fast pyrolysis system; SC, Scenario; seJ, solar emJoule or equivalent solar energy; %Renewable, Renewable fraction of total emergy in terms of percentage; NREL, National renewable energy laboratory; COD, Chemical oxygen demand; SF, solid fraction; LF, liquid fraction.

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contains an ample amount of energy. Over the years, several stillage utilization techniques have been developed to recover energy and process water: (1) evaporation followed by neutralization with alkali [10]; (2) evaporation followed by incineration [10]; (3) solid–liquid separation followed by fermentation of liquid and drying of solid [11]; (4) solid–liquid separation followed by aerobic digestion of liquid fraction and drying of solid [12]; (5) solid–liquid separation followed by anaerobic digestion of liquid fraction and drying of solid [13–15]; and (6) solid–liquid separation followed by combined anaerobic and aerobic digestion of liquid fraction and combustion of solid [6,16,17]. The products from these different recovery methods can be used for animal feedstock (based on nutrition value and type of feedstocks), fertilizers, road building materials and energy sources.

These different stillage recovery methods require different level of inputs and have their own merits and demerits. While evaporation of the liquid fraction requires an extensive amount of energy, it could separate organic and inorganic matter contained of the stillage. However, a 100% recycle rate of recovered water through evaporation may not possible due to presence of process chemicals [18]. Anaerobic digestion might be a sustainable solution for removal of the residual organic matter in the stillage by converting it to biogas, which is readily usable as fuel for the biofuel production facility itself [6,13]. Anaerobic digestion followed by aerobic treatment can be used to remove additional organic matters and to recover process water [6,17]. Thus, anaerobic digestion of the liquid fraction of stillage followed by aerobic waste water treatment is adopted for analysis in this study to obtain benefits from biogas and recovered process water.

On the other hand, the solid fraction of stillage from the cellulosic biorefinery, which is mainly lignin, has low nutrient quality [15] and may not be used as animal feedstock. Incineration of the lignin and use of remaining ash and char as a fertilizer may not be wise economic decision. As of now the most economic use of lignin is direct combustion to produce process steam and electricity [6]. For instance, only about 2% of the 50 million tons of lignin produced annually from the pulp and paper industry is used for commercial products: most is used instead to generate process heat and electricity via direct combustion [7]. Recently, established cellulosic ethanol production facilities are also using direct combustion to recover energy from lignin. Despite a wide use of stillage for process steam and electricity production through direct combustion, increasing attention is being put on other biomass conversion technologies. There is still debate about whether lignin should be used for fuel [19] or biocomposites [20]; nonetheless, interest in biomass conversion into bio-oil and biochar through pyrolysis – specifically fast pyrolysis – have been growing in recent years [21]. Pyrolysis processes can be classified into two groups: (1) slow pyrolysis (heating rate <10 °C s^{-1}) and fast pyrolysis (heating rate >10 °C s^{-1}) [21]. While slow pyrolysis results in higher biochar yields, fast pyrolysis gives higher bio-oil yields. Thus, product yields from pyrolysis are highly dependent on process conditions, such as external heating rate, reactor temperature, and retention time [22]. Detailed kinetic models of biomass pyrolysis with experimental validation are available elsewhere [21,22].

Bio-oil is composed of polar organic compounds (75–80 wt%) and water (20–25 wt%), and has a heating value of about 16–36.3 MJ/kg depending on the types of feedstock [23,24]. Bio-oil can be upgraded into various biofuels, such as gasoline, diesel, and jet fuel, as well as bioproducts, such as bio-asphalt, carbon fiber, and chemicals for industrial and medical applications [21,25,26]. Biochar consists of about 30 wt% volatile matter and has a heating value of about 13–30 MJ/kg depending on the types of feedstock [23,27]. Additionally, biochar can be used as a soil amendment that increases soil carbon content and helps maintain soil properties [21,28]. Soil properties, such as aggregate stability, tensile strength, and subcritical water repellency are very sensitive to biomass removal from the field [29], and the addition of bio-char produced during pyrolysis can improve these properties [28]. Based on discussion so far, the solid fraction of stillage utilization either through direct combustion or fast pyrolysis could generate economic and commercial benefits. Thus, both direct combustion and fast pyrolysis were selected for analysis in this study.

In addition to the solid fraction of stillage, the unutilized fermentable sugars contained in waste water can be transformed into biogas through anaerobic digestion. This process was investigated by a previous techno-economic study [6], which reported that about 20.7 wt% of methane can be produced from sugars and other process chemicals. However, this process is still being researched. The biogas generated from this process can be used for several applications: for example, it may be transformed into biomethane, an alternative to natural gas [30]; it may be transformed into methanol [31]; or it may be combusted to produce electricity [6]. In this study, it was assumed that the biogas produced from waste water was used to generate electricity.

An overview of cellulosic biorefinery stillage utilization through direct combustion and anaerobic digestion is illustrated in Fig. 1a. Stillage utilization through fast pyrolysis, with identical waste water treatment, is shown in Fig. 1b. Hereafter, 'DCS (direct combustion system)' refers to both direct combustion of the solid fraction of stillage and to waste water treatment with anaerobic digestion. Similarly, 'FPS (fast pyrolysis system)' refers to both fast pyrolysis of the solid fraction of stillage and to identical waste water treatment. While direct combustion of stillage is an established technology and is used in techno-economic analyses of the biofuel production system [6,17], stillage utilization through fast pyrolysis is still being explored through experimental studies [7]. However, these experimental studies on the fast pyrolysis of lignin using different feedstocks, such as corn stover [32], wheat straw [33] and hardwood [34,35], have shown that fast pyrolysis of lignin is technically feasible. Thus, both stillage utilization methods (Fig. 1) are technically feasible. However, it is unclear which is more sustainable in terms of creating environmental stress and in energy transformation efficiency. This is an important gap in knowledge, as

Table 1
Stillage composition of cellulosic biorefinery with a production capacity of 231 million liter ethanol/year (Extracted from previous study conducted by National Renewable Energy Laboratory (NREL) [6]).

Component	Units	Stillage
Insoluble solids (IS)	%	5.90%
Soluble solids (SS)	%	6.20%
Temperature	°C	47
Pressure	atm	6.3
Ethanol	kg/hr	184
Water	kg/hr	341,765
Glucose (SS)	kg/hr	521
Xylose (SS)	kg/hr	1062
Other sugars (SS)	kg/hr	2175
Sugar oligomers (SS)	kg/hr	1612
Organic soluble solids (SS)	kg/hr	16,420
Inorganic soluble solids (SS)	kg/hr	2610
Acetic acid	kg/hr	58
Furfurals	kg/hr	862
Other organics	kg/hr	1400
Cellulose (IS)	kg/hr	1255
Xylan (IS)	kg/hr	423
Other structural carbohydrate (IS)	kg/hr	96
Lignin (IS)	kg/hr	12,475
Protein (IS)	kg/hr	3445
Cell Mass (IS)	kg/hr	944
Other insoluble solids (IS)	kg/hr	4581
Total flow	kg/hr	391,888