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.

Keywords: cellulosic biofuels, stillage utilization, direct combustion, fast pyrolysis, emergy sustainability index

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

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 liters 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 wastewater 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 waterbody or soil. Nevertheless, the stillage 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 wastewater

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 $<10\text{ }^{\circ}\text{C s}^{-1}$) and fast pyrolysis (heating rate $>10\text{ }^{\circ}\text{C s}^{-1}$) [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 wastewater 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 wastewater 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, ‘direct combustion system (DCS)’ refers to both direct combustion of the solid fraction of stillage and to waste water treatment with anaerobic digestion. Similarly, ‘fast pyrolysis system (FPS)’ 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 biorefineries are looking for a sustainable stillage utilization method that minimizes costs and environmental impacts.

The comparative sustainability and environmental impacts of DCS and FPS can be evaluated using emergy analysis (EMA). Emergy, an abbreviation of “energy memory” or “embodied energy,” is one kind of available energy (exergy) that is required directly and indirectly to make a product or service [36]. Several previous studies have used emergy analysis to determine the sustainability of different systems such as green building design [37,38], biofuels production [39–41], cropping-grazing and agro-forestry [42,43], agricultural systems [44,45], and energy production systems [46–48].

Previous studies [49,50] have evaluated sustainability of biorefineries using Life Cycle Analysis (LCA). LCA is a very useful and common method to determine emissions and to assess the impact of a product from cradle to grave throughout its full life cycle [51]. However, like most sustainability assessment tools, it is limited in the scope of indirect and natural inputs for which it can account, as it necessarily excludes some inputs with the given systems boundary [52,53]. EMA can expand the perspective of LCA and provide the added value evaluating comprehensive donor-side assessment, specifically an estimate of the total environmental support to the examined system [53]. Emergy gets around this limitation by drawing the boundary around the entire biosphere and includes all direct, indirect and natural inputs. The direct inputs are directly consumed by the system: for instance, the solid fraction of stillage is a direct input to the boiler. By contrast, an indirect input is used earlier in the production line, as an input in the manufacturing process of a system. To continue the example, an indirect input to boiler is the coal burned to forge the steel that is used for the boiler body. The natural input is the air required to combust the solid fraction of stillage in the boiler. A previous study [51] provides critical review including detailed advantages and limitations of both LCA and emergy analysis. Additionally, Buonocore et al. [54] compares the results of both LCA and emergy analyses of a geothermal power plant system.

The emergy technique allows direct comparison of different inputs by transforming all inputs into a common unit: the solar emJoule (seJ). This is done by multiplying an input by its “transformity,” which is the equivalent amount of solar energy required to produce a unit of the particular input. As all energy on earth originates as solar energy (excepting crustal heat and tidal action, for which there are conversions [55]; and energy from nuclear fission, which is

small compared to other sources), this is an accurate representation of all inputs in energetic terms. Emergy therefore expresses the amount of solar energy that is used by the biosphere, directly and indirectly, to make a product. Transformity is often expressed in terms of ‘seJ/J’ (amount of solar energy required to produce one Joule of product), but other forms of transformity such as ‘seJ/g’ (amount of solar energy required per unit gram material output), and ‘seJ/\$’ (amount of solar energy required per unit dollar products or services output) can also be used to convert products or services into equivalent solar energy. Further, emergy input to a system can be broadly divided into renewable resources, non-renewable resources, purchased resources, and services. More details on each category are included in the discussion section of this paper; detailed discussion of the overall concepts and methods of emergy analysis are available in previous studies [36,56].

Past analyses of biogas production [47], ethanol production [39], and different agricultural production methods [45] demonstrates that EMA is suitable for the analysis of long term sustainability due to its ability to holistically assess the environmental impact [57], economic costs, and economic outputs of energy production and agricultural systems. It also allows a meaningful comparison among alternative systems due to its ability to transform all inputs into the common unit of solar emergy. In this study, two equally competitive cellulosic biorefinery byproduct utilization methods, i.e., DCS and FPS, were evaluated using EMA. With EMA all diverse inputs of renewable and non-renewable energies, as well as economic goods and services, were compared for both methods. Both DCS and FPS require different level of inputs from natural resources, non-renewable resources, and services and it is essential to assess sustainability of both systems before commercial deployment. Thus, the main objective of this

study was to assess and compare the sustainability and environmental impacts of fast pyrolysis and direct combustion systems of lignin utilization (Fig.1).

2. Methods

2.1 Overview

The stillage from a theoretical cellulosic biorefinery with an annual production capacity of 231 million liters (61 million gallons) of ethanol (proposed by NREL [6]) was considered in this study. The stillage mainly contains waste water, lignin, and unutilized sugars (Table 1). As discussed earlier, this study was focused on EMA of stillage transformation into valuable products through DCS and FPS. The EMA was accomplished through the execution of three specific tasks. First, the quantity of products and services used as inputs to the system were estimated based on process models of DCS and FPS. The process models were developed using the modeling software SuperPro Designer [58]. Second, emergy diagrams (Figure 1) and tables (Table 2 and 4) were prepared based on the direct and indirect inputs identified from the process model, and utilizing published transformities (Table 3). Finally, emergy ratios and indices were calculated (Table 7) to assess the sustainability of both stillage utilization methods. Sensitivity and risk analyses were carried out on both inputs and transformities. The detailed methodology of this study is presented in the following section.

2.2 Inputs and products estimation

The quantity and composition of stillage obtained from a previous study [6] was a main input to both stillage utilization systems, i.e., DCS and FPS. In addition to stillage, additional direct and indirect inputs required to transform stillage into useful products were estimated using process

flow models. Separate process flow models for direct combustion and fast pyrolysis were developed in SuperPro Designer software v9.0009.2100 [58]. Mass and energy balance analysis as well as economic analysis was conducted using in-built functions of the SuperPro Designer. Additional details of the modeling software can be found elsewhere [58]. Data gathered from existing literature [6,7,35,33,34,17,32] was used during the modeling process to conduct mass and energy balance analyses. Generally, these data include operating parameters data and equipment data. The operating parameters data includes operating temperature, time, pressure, conversion factors, and operating labor and utilities requirements. The equipment data includes purchase price, size and dimensions, insurance and maintenance requirements. The models were developed by completing the following three steps; inputs and products were then estimated.

1. A complete set of equipment and accessories required for direct combustion of the solid fraction of stillage (mainly lignin) and for waste water treatment, along with technical specifications and purchasing prices, was gathered from previous techno-economic studies [6,17]. The same was done for fast pyrolysis of lignin [25]. All equipment and accessories associated with waste water treatment were identical for both DCS and FPS. The parameters associated with each modeling unit within the software itself were modified based on the collected data. Lastly, current prices (2015 price) were estimated for all equipment and accessories using the current price index included in the modeling software.
2. After preparing a list of required equipment and their specifications, data regarding the operating conditions of each unit procedure, along with conversion factors of inputs into products and byproducts for DCS were directly adapted from previous techno-economic

studies [6,17]. While the DCS system is already commercialized and commercial scale production data are available, FPS is still in the research phase. Thus, the operating parameters of FPS, such as temperature, processing time and conversion factors (specifically lignin into bio-oil, bio-char, and non-condensable gases) were gathered from previous experimental studies [7,35,33,34,32]. These data were key parameters for mass and energy balance analyses during the modeling process.

3. Finally, two separate process models were developed based on collected equipment and operational data. While a complete model of DCS is already available (NREL [6]), the replication of that model in the SuperPro Designer modeling software provided a common framework to estimate inputs and outputs of both systems, i.e., DCS and FPS.

After completion of mass and energy balance analysis, annual inputs and products associated with both systems were estimated in terms of energy (Joules), mass (grams) or money (\$) (Table 2 and supplementary document S1.1-1.9), and then converted into terms of per metric ton (t) stillage input. The annual stillage input ($3.10\text{E}+06$ t) was estimated by multiplying stillage input per hour (Table 1) by annual biorefinery operating hours, which was assumed to be 7920 h considering 330 days/year and 24 h/day. Because the wastewater treatment process was the same for each system, differences among inputs were only associated with the different lignin utilization processes.

2.3 System diagrams

Based on the inputs to the stillage conversion processes identified during the modeling step, energy system diagrams for direct combustion and for fast pyrolysis of lignin were prepared

using energy systems symbols [59] (Fig. 1). In this study, a boundary was used to indicate the waste treatment section of a cellulosic biorefinery, which includes recovery of waste lignin into energy products as well as waste water treatment (Fig .1).

2.4 Solar transformity of inputs and emergy analysis tables

Solar transformity is a key element of emergy analysis that is used to transform system inputs into solar energy equivalents [36]. Transformity is a measurement of the quality of a type of energy. This follows from the observation that energy transformations are hierarchical in nature, with large amounts of diffuse energy being used (degraded to low-exergy heat) to create smaller amounts of higher-concentration energy [36]. For example, many joules of sunlight are used to make comparatively fewer joules of biomass. Within a particular system, it is generally assumed that substances with a higher transformity are of higher quality: that is, they are better able to feedback and support the system [36,56]. However, the particular transformity of a substance depends on the system used to produce it. For instance, the transformity of electricity generated from natural gas will be different than the transformity of electricity generated by a nuclear power plant. Additionally, the transformity of corn stover grown with organic farming methods will be different from that grown with conventional farming methods.

Thus, a careful consideration is required to gather or assess transformity of inputs from literature specifically types of system and feedstocks used. For instance, a system under investigation uses electricity from coal based thermal power plant then choice of transformity of electricity obtained from the same system is required than transformity of electricity from hydropower, nuclear plant or renewable systems, such as solar and wind power plants. In this study, the most

appropriate values of transformities for all inputs were collected from the literature (Table 3) and variations were accounted for through uncertainty analysis [45]. Thus, minimum, mean and maximum values of transformity were determined based on previous studies [55].

In addition to these general methods, two important assumptions were made to estimate the transformity of stillage in particular. First, stillage was separated into a solid fraction that mainly contains lignin, and a liquid fraction that contains water with unutilized sugars and process chemicals (Table 1). The source of the solid fraction of stillage is corn stover. The stillage is a waste product of the cellulosic biorefinery; however, it has some economic value due to the presence of lignin and unutilized sugars. Thus, an equivalent transformity of corn stover was used to calculate a transformity for the solid fraction of stillage. The equivalent transformity was calculated in the following three steps.

1. An equivalent transformity of corn stover was estimated by dividing the transformity of corn grain by the ratio of corn stover mass to corn grain mass (0.81) [1,64].
2. An equivalent mass of stillage (1.63 kg stillage ~ 1kg corn stover) was estimated based on the heating value of dry corn stover [75] and of stillage with a moisture content of 45%. To estimate the heating value of stillage, the heating values of cellulose (17 MJ/kg), hemicellulose (16.63 MJ/kg), and lignin (21.13 MJ/kg) [75], and their corresponding mass fractions (Table 1) were used.
3. The transformity of stillage (seJ/g) was estimated by dividing the transformity of corn stover by the calculated equivalent mass of stillage.

The above procedure effectively assumes that, by using stillage to create value-added products, an equivalent amount of corn stover must be produced to be combusted in its place. On the other hand, a published transformity for well water was used for the liquid fraction of stillage. Well water was assumed as the source of water for the previous techno-economic study of a biofuels production facility [6] upon which the stillage input values used in this paper were based. In addition to water (about 95%), the liquid fraction of stillage contains unutilized sugars and chemicals (about 5%) as summarized in Table 1 [6]. The energy input from these chemicals was included alongside the energy from well water in calculating the total energy content of wastewater. A separate transformity of those process chemicals and sugars was not considered in this study to avoid double-counting.

The commercial process model developed in the previous step provided the quantities of inputs and outputs (Table 2). Those inputs were classified as renewable resources, non-renewable resources, purchased resources, and services for both DCS and FPS, respectively. These inputs were aggregated using the standard energy classification of renewable and non-renewable energy [76]. Other non-renewable energies and other goods brought in from outside the system boundary are classified as purchased materials. Finally, purchased services include any direct and indirect services required to operate the system. Apart from these standard practices, two major assumptions were made during the input estimation. First, the total electricity input (Table 2) was divided into two groups (Table 4): (1) renewable fraction (6.82%) and (2) non-renewable fraction (93.18%), based on the mixture of renewable and non-renewable resources used for electricity production in the US [77]. Second, the solid fraction of stillage (Table 2) was categorized into three groups (Table 4): (1) renewable (14%); (2) non-renewable (4%); and (3)

purchased (82%); based on renewable, non-renewable and purchased emergy inputs required for producing corn stover [45,64]. This was used because corn stover was the source of stillage in this study. This assumption suggests that the same amount of emergy is required for the production of lignin, i.e., the solid fraction of stillage, as for the production of corn stover, because corn stover is the source of the lignin [56].

Next, the values of inputs were converted to solar emergy by multiplying by the respective transformity (Table 3) (equations 1, 2, and 3). The mean value of these transformities was used for the base case estimate. Finally, emergy flow due to each input was estimated for both DCS and FPS (Table 4). In this study, the aggregated emergy of each product was considered as the net product energy. Thus, the net product transformity was defined as the ratio of the total emergy of the system to the net product energy (Table 5). This provides a more useful means of comparing the output of the two processes. Detailed calculations of each input and product transformity are provided in the footnotes of the respective tables and in supplementary document S.1.

$$\text{Energy transformity} = \frac{\text{Solar emergy (seJ)}}{\text{Energy (J)}} \dots \dots \dots (1)$$

$$\text{Mass transformity} = \frac{\text{Solar emergy (seJ)}}{\text{Gram (g)}} \dots \dots \dots (2)$$

$$\text{Dollar transformity} = \frac{\text{Solar emergy (seJ)}}{\text{Money (\$)}} \dots \dots \dots (3)$$

2.5 Emergy signature and aggregated emergy diagrams

An emergy signature diagram shows the relative contribution of each input in terms of seJ. The signature diagrams were prepared in log-scale after emergy evaluation and taking all emergy

inputs into consideration (Fig. 2). The log-scale provides visual illustration even with the large differences in magnitude among emergy inputs. Furthermore, the total value of emergy from each main category (Table 4), i.e., renewable resources, non-renewable resources, and purchased materials and services, were calculated. Emergy from air and the renewable fraction of electricity and stillage were considered as renewable resources (R). The stillage is a byproduct of corn stover: thus, the renewable fraction of resources associated with corn production was considered a renewable input and the non-renewable fraction of resources associated with corn production was considered as a non-renewable input (N). All other input materials were considered as inputs of purchased materials (M). Purchased services (S) included maintenance, tax, and insurance required for plant machinery and equipment. Purchased materials and services were pooled as “purchased emergy” (F) for the calculation of emergy indices. Lastly, emergy yield was calculated as the sum of all emergy inputs. Based on these aggregated emergy values, the aggregated emergy diagrams (Fig. 3) were prepared for both DCS and FPS.

2.6 Calculation of emergy indices

Various emergy indices, including the emergy yield ratio, the environmental loading ratio, the emergy investment ratio, the emergy sustainability index, and the percent renewable, were calculated in order to compare the sustainability of DCS and FPS (equations 4-8 [78]). The different parameters of equations 4-8 are illustrated in Fig. 3.

$$\text{Emergy yield ratio (EYR)} = \frac{\text{Emergy yield (Y)}}{\text{Purchased emergy (F)}} \dots \dots \dots (4)$$

$$\text{Environmental loading ratio (ELR)} = \frac{\text{Purchased emergy (F)} + \text{Non-renewable emergy (N)}}{\text{Renewable emergy (R)}} \dots \dots \dots (5)$$

$$\text{Emergy investment ratio (EIR)} = \frac{\text{Purchased emergy (F)}}{\text{Renewable emergy (R)} + \text{Non-renewable emergy (N)}} \dots \dots \dots (6)$$

$$\text{Emergy sustainability index (ESI)} = \frac{\text{Emergy yield ratio (EYR)}}{\text{Environmental loading ratio (ELR)}} \dots \dots \dots (7)$$

$$\text{Fraction renewable (\%R)} = \frac{\text{Renewable emergy(R)}}{\text{Emergy yield (Y)}} \dots \dots \dots (8)$$

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388 **2.7 Sensitivity, uncertainty and scenario analysis**

389 In order to evaluate the relative impact of each input on the different emergy indices, a
 390 sensitivity analysis was performed. During sensitivity analysis all emergy inputs were doubled
 391 and halved from the average value [47]. Because there is variability in the transformities
 392 reported in the literature, a second sensitivity analysis was performed by varying the
 393 transformities of inputs ($\pm 20\%$). While sensitivity analysis only shows the relative impact of
 394 each input transformity on the outputs, uncertainty analyses for ESI and for net product
 395 transformity were carried out to determine the impact of random variation in all transformities at
 396 one time. Thus, minimum, mean and maximum values (Table 3) of all the collected
 397 transformities for each input were estimated. With the limited available data, a triangular
 398 probability distribution was assumed for all input transformities (supplementary document S.3)
 399 and 5000 random values drawn from these distribution were used for the analysis [79].
 400 Furthermore, three different scenarios were evaluated (Table 8). During the base case, the
 401 average value of each transformity was assumed, as well as the average renewable and non-
 402 renewable fraction of electricity and of the solid component of stillage. Scenario 1 (SC1)
 403 assumed a more sustainable corn production system, resulting in a higher renewable fraction for
 404 the solid component of stillage. Scenario 2 (SC2) partitioned labor into renewable and non-
 405 renewable fractions, which is a common practice in emergy analysis [45]. Finally, scenario 3
 406 (SC3) evaluated the fast pyrolysis system without electricity generation to observe the impact of

the gas compressor and turbine based electrification system on the overall sustainability of the process and to suggest alternative use of all available combustible gases.

3. Results and discussion

As discussed above, all direct and indirect material and service inputs to DCS and FPS (Table 2) were divided into four categories (Table 4): renewable resources, non-renewable resources, purchased materials, and services. Like most industrial processes, the systems investigated in this study are highly dependent on purchased resources. Emergy flow from each of those categories and evaluation of indices are discussed in the following section.

3.1 Renewable and non-renewable resources

Renewable resources account for an emergy input of $3.35\text{E}+13$, and $3.42\text{E}+13$ seJ/t stillage for DCS, and FPS, respectively. Oxygen (in air) and the renewable fraction of electricity and of solid stillage were the only renewable inputs to both systems (Table 4). Among these renewable resources, the renewable fraction of stillage was the major contributor, followed by oxygen and the renewable fraction of electricity. The renewable fraction of stillage contributed 51% and 50% of the total renewable emergy for DCS and FPS, respectively. In fact, both systems have the same amount of renewable fraction of stillage, as both have the same input of stillage: the small difference in percentage is due to the difference in the total renewable emergy between DCS and FPS. Oxygen contributed 45.97 % for DCS and 32.16 % for FPS. The emergy input from oxygen for DCS was about 40 % greater than that for FPS. In these systems, oxygen is mainly required for the combustion of fuel and for the bio-oxidation process of wastewater treatment. While DCS uses both lignin and biogas generated from the wastewater treatment process as a fuel to produce

electricity, FPS only uses biogas. Additionally, oxygen is not required for the transformation of the solid fraction of stillage into bio-oil and biochar during FPS. Thus, DCS requires 1.40 times more oxygen for the complete combustion of fuels when compared to FPS, and as a result requires a higher renewable emergy input. The balance of the renewable inputs was from the renewable fraction of electricity. FPS uses 5.67 times more electricity than DCS due to the increase in handling and processing equipment associated with this process. Apart from renewable emergy, the non-renewable fraction of stillage contributed an annual emergy input of $4.53\text{E}+12$ seJ/t stillage for both systems (Table 4).

3.2 Purchased materials and services

The major inputs of purchased materials were both the liquid and solid fractions of stillage, electricity, waste treatment chemicals, cooling water, and process steam. The purchased materials contributed an emergy flow of $3.59\text{E}+14$ and $4.50\text{E}+14$ seJ/t stillage for DCS and FPS, respectively. Wastewater, the solid fraction of stillage, waste treatment chemicals, process steam, and electricity contributed 47.91 %, 27.58 %, 11.25 %, 8.11 %, and 4.07 % of total purchased material emergy for DCS, respectively. Similarly, waste water constituted the largest input of purchased material emergy (38.22 %) for FPS followed by the solid fraction of stillage (22.00 %), electricity (18.40 %), waste treatment chemicals (8.98 %), process steam (7.51 %), and cooling water (4.56 %). All other emergy inputs of purchased materials to both systems were less than 1% of the total (Table 4). Emergy from electricity and cooling water are main differences between the purchased emergy inputs of DCS and FPS (Table 4). FPS requires 5.67 times more electricity than DCS in order to operate additional equipment required for feedstock processing (drying) as well as producing and separating biooil and biochar. Additionally, while both

systems require cooling water to maintain feedstock temperature for the anaerobic digestion of wastewater, FPS requires additional cooling water to separate bio-oil. Overall, FPS requires 9.19 times more cooling water than DCS.

Maintenance, tax, and insurance were considered as inputs of services to both systems. The total emergy contributed by services was $3.81\text{E}+13$ and $3.90\text{E}+13$ seJ/t stillage during DCS and FPS, respectively.

3.3 Emergy signature diagrams

Aggregated emergy inputs from renewable, non-renewable, and purchased resources for DCS and FPS are summarized in Fig. 2. Wastewater contributed the most emergy to both systems: 39.54 % and 32.58 % of the total emergy input for DCS and FPS, respectively. The second largest emergy input was the purchased fraction of solid stillage (i.e., lignin), accounting for 22.76 % and 18.75 % of total emergy for DCS and FPS, respectively. The waste treatment chemicals were the third largest emergy input (9.29 %) for DCS, followed by services (8.76 %), process steam (6.69 %), the renewable fraction of solid stillage (3.93 %), oxygen (3.54 %), the non-renewable fraction of electricity (3.36 %) and non-renewable fraction of solid stillage (1.04 %). On the other hand, electricity (15.68 %) was the third largest emergy input for FPS followed by waste treatment chemicals (7.65 %), services (7.39 %) process steam (6.40 %), cooling water (3.88 %), the renewable fraction of solid stillage (3.24 %), oxygen (2.08 %) and the renewable fraction of electricity (1.15 %). All other direct and indirect inputs for both systems contributed less than 1% of the total emergy input. These remaining inputs only contributed 1.14 % and 1.17 % of the total emergy input for DCS and FPS, respectively.

The above input analysis shows that both systems are highly dependent on purchased resources to transform waste stillage into valuable energy. The estimated total purchased emergy for FPS was 1.25 times more than DCS. The major differences in the purchased resource requirements are due to differences in cooling water and electricity requirements between the two systems, as discussed earlier. In addition to purchased resources, even with such a small fraction of renewable inputs, FPS uses about 2 % more renewable emergy than DCS (Table 4). While DCS uses 1.4 times more oxygen than FPS, the FPS consumes 5.67 times more electricity than DCS, which results in a higher overall renewable emergy input.

3.4 Yields and transformities

External energy is required to transform the low grade energy available in the stillage into high grade or more concentrated energy. As discussed earlier, both systems are mainly dependent on purchased resources to transform stillage into a useful form of energy, and thus both have a relatively low renewable fraction of 0.07 and 0.06 of total emergy for DCS and FPS, respectively. Even though FPS has about 2 % higher renewable emergy input than DCS, it has a lower renewable fraction of emergy because it uses 1.21 times higher total emergy than DCS. The total emergy input (emergy yield) to DCS and FPS was $4.35\text{E}+14$, and $5.28\text{E}+14$ solar emJoules (seJ)/t stillage, respectively. Electricity, process steam, and treated water were the major outputs of DCS. The estimated transformities of these products were $2.07\text{E}+06$, $1.09\text{E}+06$, and $7.22\text{E}+07$ seJ/J, respectively (Table 5). The products of FPS were electricity, bio-oil, bio-char, and treated water. The corresponding transformities of these products were $6.20\text{E}+06$, $6.82\text{E}+05$, $4.57\text{E}+05$, and $8.06\text{E}+07$ seJ/J, respectively (Table 5). The net product transformity was $7.06\text{E}+05$, and $2.61\text{E}+05$ seJ/J for DCS and FPS, respectively (Table 5).

Among comparable systems, a lower transformity of the final product implies an increase in system efficiency, as this system then requires less energy (fewer inputs or less energy-intensive inputs) to make the product than the other system with a greater transformity. Both DCS and FPS transform low value or waste stillage into high value products through the investment of different forms of direct and indirect inputs (Table 4). Electricity and treated water are common products of both systems, and so can be used to compare the efficiencies of the different systems. The treated water is considered to be a product because it can be used as process water in upstream processes within the biofuels production system. The higher transformity of electricity and treated water from FPS when compared to DCS indicates that the fast pyrolysis system is less efficient at producing electricity and process water than DCS. Process steam is a product of DCS that is not shared with FPS, whereas bio-oil and bio-char are the other main products of FPS. Given that the two systems produced different outputs, and because of the ambiguous relationship between the transformities of the common products, a net product transformity of both systems was calculated in order to compare the overall relative sustainability of systems. A lower net product transformity in either form (i.e., seJ/J or seJ/\$) indicates that FPS is, in general, more efficient at producing economic value than DCS. This highlights an important point: that efficiency is defined relative to products: while DCS is more efficient than FPS at producing electricity and treated water, the opposite is true for the consideration of all products. Additionally, a system with the lower net product transformity and the higher energy sustainability index discussed in the following section may be the more economic system as this system requires fewer or lower energy inputs.

Table 6 summarizes the comparison among product transformities of DCS, FPS, and other systems. As no emergy analyses have been performed on identical systems involving stillage utilization, the transformities for products from both DCS and FPS were compared with products from similar systems involving coal, and other municipal and industrial wastes. The transformities of products found in this study were higher than those from previous studies (Table 6), which is mainly due to the difference in the energy contents of the input materials (i.e., feedstocks). While the system in this study attempts to transform waste into valuable products, the feedstocks for the other studies were either biomass or high-quality non-renewable fuels. These feedstocks contain high input energy value when compared to stillage and therefore can produce energy outputs, such as electricity, with fewer inputs. Additionally, the overall conversion processes used in this study are very different from those used in systems considered by the previous studies, even though the product outputs are identical. For instance, the liquid fraction of stillage can be used as a feedstock for a biodigestion system instead of livestock manure [47] or dairy waste [65] to produce first biogas and then electricity; however, the systems in this study used anaerobic digestion followed by wastewater treatment to recover waste water in the form of process water. Apart from waste water, the solid fraction of stillage was used to produce bio-oil and bio-char in FPS and electricity and process steam in DCS. Thus, the product transformities estimated in this study were higher than other energy systems with livestock manure, dairy waste and coal because of the differences in feedstocks and in processes.

3.5 Emergy ratios and indices

In addition to product transformities, five different emergy ratios and indices were calculated in order to evaluate the sustainability and environmental impact of both DCS and FPS (Table 7).

544 First, DCS was found to have a higher emergy yield ratio and a higher renewable fraction when
545 compared to FPS (Table 7). EYR measures the fraction of emergy that needs to be purchased
546 from outside the system in order to utilize local renewable emergy from within the system. Even
547 to utilize renewable resources as an input energy, some kind of high grade energy from outside
548 the system is required. For instance, to use oxygen as an input to the system, a fan, blower, or
549 compressor is required, and these are operated with high grade purchased electrical energy. With
550 only a slight difference between the EYR of DCS and FPS it was concluded that the systems had
551 a similar ability to capture locally available renewable resources. Second, the FPS has a higher
552 emergy investment ratio (EIR). The EIR measures the ratio of emergy invested into the system
553 from outside (that is, from the economy) to the locally utilized renewable energy [48]. Thus, FPS
554 is less economically efficient, in emergy terms, in that it requires about 23.17 % more invested
555 resources to use locally available resources when compared to DCS. Third, FPS has a higher
556 environmental loading ratio (ELR) than DCS. The ELR measures the ratio of purchased and non-
557 renewable resources used to the renewable resources used; the idea is that the non-renewable
558 emergy added to the system has the potential to cause environmental stress by overwhelming the
559 system, but only to the degree in which this exceeds the amount of emergy that is naturally
560 present in the system [78]. Thus, FPS showed a higher potential for causing environmental stress
561 than DCS. Fourth, the estimated emergy sustainability index (ESI) of DCS was 22.67% higher
562 than that for FPS. The ESI measures the sustainability of the overall system by measuring the
563 contribution of a resource to the economy per unit of environmental loading [78]. The lower ESI
564 of FPS indicates that FPS is less sustainable than DCS. One of the major reasons for this is that
565 FPS uses more purchased inputs, specifically electricity and cooling water, in order to produce
566 electricity, process steam, process water, bio-oil and bio-char. FPS also has a much lower

requirement of oxygen for combustion than DCS: 8.06 kg/s instead of 18.03 kg/s (supplementary document S1.1): this is because it has less fuel available and more importantly creates less electricity. However, there was only a slight difference between the ESI of both systems. Additionally, there is ample space for process optimization of FPS including improved operation temperatures and times, and an improved lignin to bio-oil and bio-char conversion rate as well as process intensification, such as more efficient compressors and blowers. These improvements could potentially make the sustainability of FPS comparable to that of DCS.

3.6 Sensitivity, scenario, and uncertainty analysis

A sensitivity analysis on ESI, ELR, and net product transformity of both FPS and DCS was carried out to assess the impact of variation in inputs and transformities and to identify the most significant contributors to these indices (Fig. 4).

For sensitivity analysis, all inputs were doubled and halved. The ESI for both systems was most sensitive to changes in the emergy contributed by the renewable fraction of stillage followed by oxygen, wastewater and the purchased fraction of stillage (Fig. 4). This makes sense, as these first two were the largest renewable inputs and last two were the largest purchased inputs to both systems (Fig.2). Increasing renewable inputs and decreasing purchased inputs increases ESI, thus the sensitivity results also explain in part the higher ESI for DCS: as discussed in the previous section, DCS has a higher renewable and lower purchased fraction of emergy than FPS. Apart from those input parameters, waste treatment chemicals was found to be the fifth most sensitive input for DCS, followed by services, process steam, the non-renewable fraction of electricity and the renewable fraction of electricity. For FPS the renewable fraction of electricity

was the fifth most sensitive input followed by the non-renewable fraction of electricity, wastewater treatment chemicals, services, process steam and cooling water. ESI was not very sensitive to variations in all other inputs. Additionally, ESI showed a similar sensitivity to $\pm 20\%$ variation in transformity of all inputs (Fig. 5). An analysis on the sensitivity of ELR to different inputs found similar results (supplementary document S2). In contrast, the net product transformity was most sensitive to purchased inputs including wastewater and the purchase fraction of stillage, respectively (supplementary document S2). Overall, the sustainability of FPS is more sensitive to the renewability of the corn farming process. Therefore, if the corn stover is produced in a more sustainable way, FPS will benefit more than DCS.

In addition to the sensitivity analysis done on the base case results, the three different scenario analyses (Table 8) also indicate that DCS is more sustainable than FPS. Table 8 summarizes the energy ratios and indices under three different scenarios and the base case scenario. The highest ESI value for FPS (0.104) was obtained for the scenario without electricity generation (SC3); this is more than the base case ESI value of 0.09 for DCS (Table 7). On the other hand, ESI values of 0.13 and 0.10 were found under the first scenario (SC1) (Table 8) for DCS and FPS, respectively. Finally, results from second scenario (SC2) were found similar to the base case scenario.

In each scenario a significant change in the renewable fraction was found. When the renewable fraction of solid stillage was increased by 9 % (assuming more sustainable corn production) (SC1), the ESI value of FPS and DCS increased by 47.1 and 42.2 %, respectively. This result indicates that the renewable fraction of stillage is more influential for FPS than for DCS. Labor

was found to be non-influential in the sensitivity analysis; thus, assigning renewable and non-renewable fractions to the input of labor (SC2) did not significantly change the ESI value or any other ratios for either system. Finally, FPS without electricity generation (SC3) was evaluated to observe the impact of the number of compressors required for electricity production. While DCS uses a steam turbine-generator to produce electricity, where the required pressure is achieved with a water pump to feed water to the boiler, the electricity production system of FPS assumed in this study requires the use of a gas turbine-generator system, which required several compressors to achieve the necessary maximum pressure of 6 bar. Besides the expense of materials, these compressors also consumed a significant amount of electricity. Thus, electricity generation might not be economically or environmentally feasible for the FPS process. In this scenario, the purchased inputs for FPS were decreased and the ESI value of FPS became very close to the ESI value of DCS in the base case. In this case, FPS is more competitive with DCS. A similar trend was found in a previous study [47] in which a biodigestion system was evaluated with and without electricity production from biogas. Thus, the ESI value of both stillage recovery systems could be improved by increasing renewable energy inputs via a more sustainable corn production system, and by improving system efficiency to reduce requirements for purchased electricity, cooling water and steam.

Furthermore, Fig. 6 illustrates the uncertainty analysis on the ESI value of both DCS and FPS. The uncertainty analysis showed that a 95% confidence interval for ESI was 0.07-0.21 and 0.05-0.16 for DCS and FPS, respectively. This indicates a higher uncertainty in the ESI value of DCS when compared to that of FPS. Similar results were found for the uncertainty analysis on net product transformity (Fig. 7). However, most of the estimated ESI values of both systems

overlap each other suggesting that the choice between these systems depend largely upon the desired end products and end uses.

4. Conclusion

This study estimated a slightly higher value of the emergy sustainability index for the direct combustion system, providing it some advantage over the fast pyrolysis system in terms of emergy sustainability. At the same time, the net product transformity of the fast pyrolysis system is lower than that of the direct combustion system due to its multiple output products, providing it some advantage over the direct combustion system in terms of emergy utilization efficiency. The emergy sustainability index could be improved and the product transformity could be reduced for both the systems by decreasing purchased inputs; however, in this respect there are more opportunities with fast pyrolysis process as it is still at a very early stage of process development and commercialization when compared to the direct combustion system. The purchased inputs can be decreased via process optimization and intensification, which are promising directions for future research. Process optimization includes the choice of optimum input and process parameters under the current system, and process intensification includes a re-design of the current system with alternative good technologies. For instance, purchased inputs of fast pyrolysis could be reduced with the optimum pyrolysis reactor temperature and time, as well as the use of a gas engine-generator system instead of a gas turbine-generator system. Likewise, an energy efficient boiler and steam turbine could reduce purchased inputs for direct combustion. Additionally, a more effective solid-liquid separation unit that can produce solid cake with low moisture content could improve sustainability of both the systems due to a lower

heat energy input requirement to remove moisture. Thus, both systems currently have similar levels of emergy sustainability, and both can be further improved through process optimization.

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(b) Fast pyrolysis system

Fig. 1 System diagram of cellulosic biorefinery stillage utilization through direct combustion and fast pyrolysis

(Items outside the system boundary and inscribed within circles are direct and indirect inputs of energy, materials, and services into the system. A fraction of stillage only contains waste water, which goes directly to the anaerobic digestion unit [6]. In contrast, stillage with mixture of waste water and lignin goes through centrifugation. The two-way arrows indicate recycle of input material. Electricity and process steam are the outputs of the direct combustion system (Fig. 2 (a)). In contrast, the major outputs of the fast pyrolysis system are electricity, bio-char, and bio-oil (Fig. 2 (b)). Some important material(s) associated with the paths as indicated in Fig. 1 (a) and (b) are: #1: waste water; #2: lignin; #3: waste water; #4: additional water and biogas; #5: sludge and treated water; #6: steam; #7: biogas; #8: dried lignin; #9: bio-oil, bio-char and combustible gases; #10: bio-oil and combustible gases; #11: combustible gases.)

(a)

(b)

Fig. 2 Emenergy signature diagrams of biorefinery stillage utilization methods: (a) Direct combustion system, and (b) Fast pyrolysis system

(In these diagrams: “Stillage SF” indicates the solid fraction of stillage (mainly lignin); “Stillage LF” indicates the liquid fraction of stillage (waste water and chemicals); “WT chemicals” refers to waste treatment chemicals; and “MTI” refers to maintenance, tax and insurance services.)

Fig. 3 Summary diagrams of energy flow for cellulosic biorefinery stillage utilization methods: (a) Direct combustion system, and (b) Fast pyrolysis system

(In these diagrams: R = renewable resources input; N = nonrenewable resources input; M = purchased materials input; S = purchased services input; F = purchased resources input = M + S; and Y = total energy flow. All energy values are in 10^{13} seJ/t stillage. Detailed calculations are presented in Table 4.)

Fig. 4 Sensitivity of energy inputs on energy sustainability index (ESI): (a) Direct combustion system; and (b) Fast pyrolysis system; <20% of the base case value and >20% of the base case value

(Average ESI was 0.092 for direct combustion (Fig. 4 (a)) and 0.075 for fast pyrolysis (Fig. 4 (b)). These figures illustrate the impact on ESI due to doubling and halving the individual energy inputs.)

(a)

(b)

Fig. 5 Sensitivity of inputs transformity on energy sustainability index (ESI): (a) Direct combustion system; and (b) Fast pyrolysis system; <20% of the base case value and > 20% of the base case value

(Average ESI was 0.092 for direct combustion (Fig. 5 (a)) and 0.075 for fast pyrolysis (Fig. 5 (b)). These figures illustrate the effect on ESI due to $\pm 20\%$ variation in transformity for each energy input.)

(a)

(b)

Fig. 6 Uncertainty analysis on emergy sustainability index (ESI): (a) Direct combustion system; and (b) Fast pyrolysis system

(After 5000 trials, the 95% certainty value of ESI was 0.21 for direct combustion (Fig. 6 (a)) and 0.16 for fast pyrolysis (Fig. 6 (b)). These figures illustrate the effect on ESI due to variation in the transformities of emergy inputs. A wider curve of DCS when compared FPS indicates higher uncertainty. The probability distribution of all inputs are provided in the supplementary document S.5.)

(a)

(b)

Fig. 7 Uncertainty analysis on net product transformity: (a) Direct combustion system; and (b) Fast pyrolysis system

(After 5000 trials, the 95% certainty value of net product transformity was 1.07 E+06 seJ/J for direct combustion (Fig. 7 (a)) and 3.75 E+05 seJ/J for fast pyrolysis (Fig. 7 (b)). The probability distributions of all inputs are provided in the supplementary document S.5.)

List of Tables

Table 1. Stillage composition of cellulosic biorefinery with a production capacity of 231 million liter ethanol/year (Extracted from previous study 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	1,062
Other sugars (SS)	kg/hr	2,175
Sugar Oligomers (SS)	kg/hr	1,612
Organic Soluble Solids (SS)	kg/hr	16,420
Inorganic Soluble Solids (SS)	kg/hr	2,610
Acetic Acid	kg/hr	58
Furfurals	kg/hr	862
Other Organics	kg/hr	1,400
Cellulose (IS)	kg/hr	1,255
Xylan (IS)	kg/hr	423
Other Structural Carbohydrate(IS)	kg/hr	96
Lignin (IS)	kg/hr	12,475
Protein (IS)	kg/hr	3,445
Cell Mass (IS)	kg/hr	944
Other Insoluble Solids (IS)	kg/hr	4,581
Total Flow	kg/hr	391,888

Table 2. Material and energy input and products from direct combustion and fast pyrolysis system

Parameter	Units	Unit/year		Unit/t stillage ¹	
		DCS	FPS	DCS	FPS
Input²					
Oxygen	g	9.41E+11	6.73E+11	3.04E+05	2.17E+05
Stillage (biomass based)	g	1.13E+11	1.13E+11	3.65E+04	3.65E+04
Stillage (waste water) ³	J	2.88E+15	2.88E+15	9.29E+08	9.29E+08
Additional process water	g	1.17E+12	1.17E+12	3.77E+05	3.77E+05
Cooling water ⁴	J	1.16E+14	1.07E+15	3.74E+07	3.45E+08
Process steam ⁵	J	1.37E+15	1.59E+15	4.42E+08	5.13E+08
Electricity	J	2.74E+14	1.55E+15	8.84E+07	5.00E+08
Waste treatment chemicals	g	1.79E+10	1.79E+10	5.77E+03	5.77E+03
Equipment steel	g	1.38E+07	2.55E+07	4.45E+00	8.23E+00
Reinforcing steel	g	7.08E+07	7.08E+07	2.28E+01	2.28E+01
Cement	g	8.01E+07	8.01E+07	2.58E+01	2.58E+01
Labor	h	2.98E+05	2.81E+05	9.61E-02	9.06E-02
Operating cost (maintenance, tax, & insurance)	\$	2.27E+07	2.32E+07	7.31E+00	7.49E+00
Product⁶					
Electricity	J	6.52E+14	2.64E+14	2.10E+08	8.52E+07
Process steam	J	1.24E+15	-	4.00E+08	-
Process water	J	1.87E+13	2.03E+13	6.03E+06	6.55E+06
Bio-oil	J	-	2.40E+15	-	7.74E+08
Bio-char	J	-	3.58E+15	-	1.15E+09
Net product	J	1.91E+15	6.26E+15	6.16E+08	2.02E+09

¹Total annual stillage from 231 million liter annual ethanol production facility was 3.10E+06 metric ton (t)) (Table 1) assuming biorefinery operating hours of 7920 h (330 days/year and 24 h/day).

²Detail estimation of inputs are provided in the supplementary document. Unless otherwise stated these inputs were obtained from techno-economic model developed in modeling software- SuperPro Designer assuming biorefinery operating hour of 7920 h (330 days/year and 24 h/day).

³The energy contained in wastewater includes energy of both water and dissolved chemicals. The energy contained in water was assumed to be 4.91 J/g [46]. The energy contained in chemicals was assumed to be 14233.4 J/g COD [46]. The estimated COD value of wastewater was 25.43metric ton (t)/h.

⁴Energy contained in the cooling water was assumed to be 0.0058 kWh/kg [58], (1 kWh = 3600000 J).

⁵Energy contained in the process steam water was assumed to be 0.536 kWh/kg [58].

⁶Products were obtained from techno-economic model developed in modeling software- SuperPro Designer (supplementary document-S1.9). Energy value of electricity, process steam and process water were directly obtained from process model. Additionally, energy conversion factor for bio-oil and biochar were 24.3 MJ/kg and 33.3 MJ/kg, respectively (1MJ = 1E+06 J) [27].

Table 3. Transformities of various inputs

Parameter	units	Transformity ¹			References
		Minimum	Mean	Maximum	
Oxygen	seJ/g	4.95E+07	5.06E+07	5.16E+07	[55,60–63]
Stillage (solid)	seJ/g	9.14E+08	3.31E+09	1.02E+10	[56,45,64]
Stillage (liquid)	seJ/J	6.60E+04	1.85E+05	2.45E+05	[65–67]
Additional process water	seJ/g	3.27E+05	1.99E+06	5.09E+06	[60,68,69,37]
Cooling water	seJ/J	5.83E+04	5.95E+04	6.07E+04	[70]
Process steam	seJ/J	6.45E+04	6.59E+04	6.72E+04	[70]
Electricity	seJ/J	1.14E+05	1.78E+05	2.58E+05	[57,60,65,37,71]
Waste treatment chemicals	seJ/g	6.86E+09	7.00E+09	7.14E+09	[39,65,46]
Cement	seJ/g	8.89E+07	4.87E+09	3.18E+10	[39,60,65,69,37,46]
Reinforcing steel	seJ/g	3.19E+09	4.75E+09	6.69E+09	[39,57,69,37,71,46,72,73]
Equipment steel	seJ/g	1.71E+09	6.33E+09	1.25E+10	[56,39,60,73,74]
Labor	seJ/h	3.80E+12	6.53E+12	8.45E+12	[45,65,67]
Maintenance, tax, & insurance	seJ/\$	1.06E+12	5.21E+12	1.16E+13	[39,65,67,72]

¹ All transformities were calculated based on the latest global emergy baseline value of 15.2E24 seJ/year [55].

Previous values of this baseline were 15.83E24 seJ/year [74], and 9.44E24 seJ/year [56].

Table 4. Emergy inputs, transformity, and total emergy required for stillage utilization through direct combustion and fast pyrolysis system

Index	Input	Units	Value (unit/t stillage)		Transformity (seJ/unit)	Emergy (seJ/t stillage)	
			DCS	FPS		DCS	FPS
Renewable resources (R)							
1	Oxygen	g	3.04E+05	2.17E+05	5.06E+07	1.54E+13	1.10E+13
2	Electricity (renewable)	J	6.04E+06	3.42E+07	1.78E+05	1.07E+12	6.07E+12
3	Stillage (Solid fraction, renewable)	g	5.17E+03	5.17E+03	3.31E+09	1.71E+13	1.71E+13
Total renewable						3.35E+13	3.42E+13
Nonrenewable resources (N)							
4	Stillage (solid fraction, nonrenewable)	g	1.37E+03	1.37E+03	3.31E+09	4.53E+12	4.53E+12
Total non-renewable						4.53E+12	4.53E+12
Purchased resources (M)							
5	Stillage (solid fraction, purchased)	g	2.99E+04	2.99E+04	3.31E+09	9.90E+13	9.90E+13
6	Stillage (waste water)	J	9.29E+08	9.29E+08	1.85E+05	1.72E+14	1.72E+14
7	Additional process water	g	3.77E+05	3.77E+05	1.99E+06	7.49E+11	7.49E+11
8	Cooling water	J	3.74E+07	3.45E+08	5.95E+04	2.23E+12	2.05E+13
9	Process steam	J	4.42E+08	5.13E+08	6.59E+04	2.91E+13	3.38E+13
10	Electricity (non-renewable)	J	8.23E+07	4.66E+08	1.78E+05	1.46E+13	8.28E+13
11	Waste treatment chemicals	g	5.77E+03	5.77E+03	7.00E+09	4.04E+13	4.04E+13
12	Equipment steel	g	4.45E+00	8.23E+00	6.33E+09	2.82E+10	5.21E+10
13	Reinforcing steel	g	2.28E+01	2.28E+01	4.75E+09	1.08E+11	1.08E+11
14	Cement	g	2.58E+01	2.58E+01	4.87E+09	1.26E+11	1.26E+11
15	Labor	h	9.61E-02	9.06E-02	6.53E+12	6.28E+11	5.92E+11
Total purchased (M)						3.59E+14	4.50E+14
Services (S)							
16	Maintenance, tax, & insurance	\$	7.31E+00	7.49E+00	5.21E+12	3.81E+13	3.90E+13
Total services (S)						3.81E+13	3.90E+13
Total purchased and services (F) = M+S						3.97E+14	4.89E+14
Grand total emergy inputs (R+N+F)						4.35E+14	5.28E+14

Table 5. Products from stillage processing through DCS and FPS and transformity of products

Index	Product	Unit	Value (unit/t stillage)		Product transformity (seJ/unit)*	
			DCS	FPS	DCS	FPS
Outputs (Y)						
1	Electricity	J	2.10E+08	8.52E+07	2.07E+06	6.20E+06
2	Process steam	J	4.00E+08	-	1.09E+06	-
3	Treated water	J	6.03E+06	6.55E+06	7.22E+07	8.06E+07
4	Bio-oil	J	-	7.74E+08	-	6.82E+05
5	Bio-char	J	-	1.15E+09	-	4.57E+05
6	Net product	J	6.16E+08	2.02E+09	7.06E+05	2.61E+05

*Products' transformity was estimated by dividing the grand total emergy inputs/t stillage (Table 4) by the total available energy/t stillage of each product.

Table 6. Comparison of product transformity with different energy systems

Energy system/product	Transformity of product, seJ/J	Relative difference with respect to previous value (%)	References
<i>Product: Electricity</i>			
Electricity generation from stillage with FPS	6.20E+06	-	This study
Electricity generation from stillage with DCS	2.07E+06	-66.62	This study
Electricity generation from Livestock manure	1.59E+06	-23.17	Ciotala et al. [47]
Electricity generation from dairy waste	1.19E+06	-25.16	Bastianoni and Marchettini [65]
Electricity generation from coal	1.60E+05	-86.55	Odum [56]
<i>Product: Oil/bio-oil</i>			
Bio-oil from stillage	6.82E+05	-	This study
Bio-oil from sugarcane trash	6.97E+04	-89.78	Zhang and Long [60]
Oil	5.40E+04	-22.53	Odum [56]
<i>Product: Char/bio-char</i>			
Bio-char from stillage with FPS	4.57E+05	-	This study
Char	4.00E+04	-91.25	Odum [56]
<i>Product: Process steam</i>			
Process steam from stillage	1.09E+06	-	This study
Process steam from coal	6.59E+04	-93.94	Wang et al. [70]
<i>Product: Process water</i>			
Process water from stillage with DCS	7.22E+07	-	This study
Process water from stillage with FPS	8.06E+07	11.73	This study

Table 7. Emergy ratio and indices for stillage utilization through direct combustion and fast pyrolysis

Emergy indices and ratio	Direct combustion	Fast pyrolysis
Emergy yield (Y), seJ/year	4.35E+14	5.27E+14
Emergy yield ratio (EYR)	1.10	1.08
Emergy investment ratio (EIR)	10.44	12.65
Environmental loading ratio (ELR)	11.99	14.46
Emergy sustainability index (ESI)	0.09	0.07
% Renewable	7.70	6.47

Table 8. Scenario analysis of emergy indices and ratios for both direct combustion and fast pyrolysis systems

Emergy indices and ratio	Base case		SC1 ^a		SC2 ^b		SC3 ^c	
	Direct combustion	Fast pyrolysis	Direct combustion	Fast pyrolysis	Direct combustion	Fast pyrolysis	Direct combustion	Fast pyrolysis
Emergy yield (Y), seJ/year	4.35E+14	5.27E+14	4.35E+14	5.27E+14	4.35E+14	5.27E+14	-	4.11E+14
Emergy yield ratio (EYR)	1.10	1.08	1.13	1.11	1.10	1.08	-	1.10
Emergy investment ratio (EIR)	10.44	12.65	7.47	9.15	10.42	12.62	-	10.22
Environmental loading ratio (ELR)	11.99	14.46	8.86	10.79	11.96	14.42	-	10.57
Emergy sustainability index (ESI)	0.09	0.07	0.128	0.103	0.092	0.075	-	0.104
% Renewable	7.70	6.47	10.14	8.48	7.72	6.48	-	7.80

^aAssuming sustainable corn production system with renewable (R), nonrenewable (N) and purchased (F) emergy input of 23 %, 6%, and 71%, respectively [64]. The corresponding average value of R, N, and F for the base case was 14%, 4%, 82%, respectively.

^bAssuming renewable (13%) and nonrenewable (87%) fraction of labor [45]

^cFast pyrolysis system without electricity generation from biogas