Process Simulation Integrated Life Cycle Net Energy Analysis and GHG Assessment of Fuel-Grade Bioethanol Production from Unutilized Rice Straw

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Statement of Novelty for:

Process simulation integrated life cycle net energy analysis and GHG assessment of fuel-grade bioethanol production from unutilized rice straw

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Published life cycle assessments (LCA) for bioethanol production from rice straw have incorporated an inventory allocation factor for the paddy rice cultivation stage, considering mass/economic values of rice grain and straw. However, for the scenario of unutilized rice straw that remains as waste biomass with no economic value, inventory allocation for the rice cultivation stage can be excluded. The effects of this life cycle consideration have not been comprehensively interpreted in the published literature. Therefore, this study assesses the life cycle net energy analysis and GHG assessment for unutilized rice straw valorization via scaled-up bioethanol production with a zero-inventory allocation rule for the rice cultivation stage. The findings would support future LCAs for unutilized rice straw valorization processes.

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All the data sources are cited in this manuscript and additional data with their sources are included as electronic supplementary material attached to the manuscript.

Abstract

The life cycle stage of paddy rice cultivation can be excluded with a zero-inventory allocation rule for the life cycle scenario of bioethanol production from unutilized rice straw, i.e., rice straw with no applied valorization in current practice. Accordingly, this study evaluates the life cycle net energy analysis and greenhouse gas (GHG) assessment for a scaled-up bioethanol production plant using unutilized rice straw as the feedstock. The process simulation technique is integrated to model a scaled-up production plant to produce bioethanol at 99.7 vol% purity from unutilized rice straw, and the simulation results are retrieved to calculate inventory data for life cycle assessment (LCA). The simulated mass flow and energy flow results are comparable with that of real plants, reported in the published literature, which validates the process simulations in this study. Inclusive of energy generation using the waste flows in the process (i.e., wastewater and solid residues), the life cycle net energy analysis results show a net energy gain of 7,804.0 MJ/m^3 of bioethanol with a net renewable energy gain of 38,230.9 MJ/m^3 of bioethanol that corresponds to a net energy ratio of 1.20 and renewability factor of 5.49. The life cycle GHG assessment exhibits a net global warming potential of 584.8 kg CO_2 eq/m^3 of bioethanol. The effect of system boundary expansion up to the end-of-life stage as gasohol (E10), the sensitivity of the key process parameters, and the economic benefit via valorization of unutilized rice straw are further analyzed and discussed.

Keywords: Fuel-grade bioethanol production, Unutilized rice straw, Process simulation integrated LCA, Life cycle net energy analysis, Greenhouse gas assessment

1. Introduction

Global energy consumption increases along with the rapid growth of population and technology. The transportation sector consumes approximately 33% of global energy consumption and increases its energy demand at an annual average rate of 1.4% [1, 2]. Currently, this increasing demand is compensated from the major petroleum derivatives, such as gasoline and diesel. With the rapid depletion of petroleum crude oil resources, it would be a huge challenge to satisfy the rising energy demand for the next decades [3]. In addition to the future risk of fuel resources depletion, environmental impacts due to pollutant emissions from the consumption of petroleum derivatives as transportation fuels would make the problems more complicated. However, fuel blending with liquid biofuels is an attractive solution that can reduce the depletion rate of petroleum resources and the associated environmental impacts to a significant extent.

Many countries have introduced blended biofuels into their national energy policies with future goals of expanding biofuels production. Hence, a major concern has been drawn to bioethanol as a successful blending agent for gasoline. For example, the European Union has launched tangible action plans to achieve the targeted circular economy through promoting biofuels by 2030 [4, 5]. Japan has developed a criterion to expand the country's biofuel production with a long-term target of six thousand million liters of bioethanol, including lignocellulosic feedstocks [6]. A South Asian country like India has also taken steps to promote bioethanol with a mandatory blending ratio of 20% [7]. Furthermore, the major automobile manufacturers including Mercedes Benz, Toyota, etc. have also introduced Flexible Fuel Vehicles (FFV) to promote the utilization of bioethanol as a transportation fuel. Approximately five million FFVs are used currently in the USA and about 70% of new vehicle purchases in Brazil are FFVs. Thus, Bioethanol would be one of the major renewable transportation fuels for the future [8]. Gasohol, which is the acronym termed for blended gasoline with bioethanol (ethyl alcohol) has commercial variants, such as E10, E20, E80, etc. These gasohol blends ranging from E3 to E85 can be used in vehicles without any engine modifications. Gasohol blends provide an added advantage of octane number enhancement, which avoids the use of toxic octane boosters in pure gasoline, such as tetraethyl lead (TEL), Methyl tertiary-butyl ether (MTBE), and other synthetic oxygenates [9].

Bioethanol is commercially produced from starch and sugar-based feedstocks (first-generation) and lignocellulosic feedstocks (second generation). Various countries, including Brazil, the USA, China, Thailand, etc. have modified their agricultural policies and transportation fuels supply chains to produce commercial fuel-grade bioethanol using more sustainable feedstocks within the respective country. Brazil uses bioethanol as its primary transportation fuel where sugarcane, cane molasses, and excess bagasse are the major feedstocks [8, 10]. The

United States utilizes corn and corn stover as the major feedstocks to produce bioethanol [8]. Thailand produces the total bioethanol demand for gasohol blending within the country mainly using industrial cassava and cane molasses as the feedstocks [11]. For an agricultural country with limitations of arable land availability and food security competition with edible feedstocks, non-edible agro-waste residues, such as rice straw, wheat straw, corn stover, bagasse, etc. would be more economically viable feedstock options [12–15]. It is expected that the production quantities of bioethanol from second-generation feedstocks would exceed the bioethanol coming from first-generation feedstocks within the next ten years [14].

Many Asian countries, such as China, India, and Thailand annually generate approximate amounts of 75, 60, and 12 million tonnes of rice straw, respectively [16]. Sri Lanka also annually generates approximately 2.8 million tonnes of rice straw where at least a single commercial-scale bioethanol production plant or any valorization is not in practice using this significant rice straw availability [17]. Hence, for the Sri Lankan context, rice straw can be considered as an unutilized residue with no valorization in practice. Open burning of unutilized rice straw after field manuring is the main practice of farmers that would cause many environmental consequences. Thus, there is a great opportunity to establish a new fuel-grade bioethanol production plant in Sri Lanka by processing this unutilized rice straw as the feedstock. It would provide foreign currency savings from petroleum crude oil importation to the country and sustainable development by achieving the country's future renewable energy policies. For this purpose, this study focuses on performing standard energy and greenhouse gas (GHG) assessment of a commercial-scale plant design considering the life cycle of bioethanol production from unutilized rice straw in Sri Lanka.

In the fuel-grade bioethanol production life cycle, rice straw is baled and brought into the process plant, initially. Then they are crushed into pieces and pre-treated to remove the lignin content. Next, the pretreated rice straw is hydrolyzed to convert the recovered cellulose and hemicellulose into sugars, such as glucose, xylose, fructose, etc. The sugar solution is then fermented to produce dilute ethanol solution, i.e., beer solution. An azeotropic ethanol solution is obtained via distillation of the beer solution. Finally, the azeotropic ethanol solution is dehydrated to obtain fuel-grade bioethanol having at least 99.5 vol% purity. Waste streams (lignin contained solid residues and spent wash from distillation columns) generated throughout this process could be utilized to generate the process energy required for the same process. For real bioethanol production plants that are operated at a commercial scale, inventory data for their Life Cycle Assessment (LCA) could be obtained using conventional data survey techniques. However, with the absence of commercial-scale plants, conventional inventory data survey techniques are not possible to assess a scaled-up bioethanol production plant for the Sri Lankan scenario. Therefore, process simulation is utilized as the technique in this study to model a scaled-up plant to produce fuel-grade bioethanol using unutilized rice straw in Sri Lanka as the feedstock and retrieve the required process data for the LCA. Some published studies have utilized the process simulation technique to retrieve required process inventory data for LCA and validated the results by comparing them with actual plant data in the published literature [18–24]. In addition, several studies have successfully used process simulation and modeling techniques for scaling up and optimization of bioethanol production processes [19, 22, 23].

Various scenarios of bioethanol production using rice straw as a feedstock in different countries have been studied to evaluate the life cycle net energy balance. An LCA study conducted in Thailand concludes that among the valorization options for rice straw, such as co-generation of heat and electricity, bio-DME production, and fertilizer production, bioethanol production is the most environmentally benign approach [25]. Studies performed in Japan and Thailand conclude that heat and electricity produced using lignin contained solid residues and biogas from spent wash recovery are sufficient (with a surplus) to cater to the process energy requirement for bioethanol plants [26, 27]. Japan has conducted a net energy analysis for bioethanol production using high yield rice plants and obtained Net Energy Ratio (NER > 1) and Renewability (Rn > 1) [26]. Further, some existing LCA studies in Thailand and India have reported both positive and negative Net Energy Values (NEV) and Net Renewable Energy Values (NRnEV) for bioethanol production from rice straw [7, 23]. In all the published studies in the literature, an allocation rule has been considered to account for the life cycle inventory data of the paddy rice cultivation stage. However, for a scenario like in Sri Lanka where valorization is not in practice for rice straw (unutilized rice straw), the inventory data of the paddy rice cultivation stage can be excluded with a zero-allocation rule. Hence, the assessment of fuel-grade bioethanol production life cycle using unutilized rice straw with zero inventory allocation from the feedstock cultivation stage would contribute a special scenario for future LCA studies.

As such, this study proposes a simulated fuel-grade bioethanol production plant from unutilized rice straw in Sri Lanka as the feedstock with life cycle net energy analysis and GHG assessment at zero inventory allocation for the feedstock cultivation stage. The economic benefit of the utilization of rice straw for fuel-grade bioethanol production with system expansion of gasohol blending as the end-of-life stage is also covered in this study. In addition to the Sri Lankan context, the findings from this study will support the scenario of bioethanol production using unutilized rice straw for any location in other parts of the world where a new plant is expected to be established.

2. Materials and Methods

2.1 Life cycle scope and process description

In this study, the process simulation technique is used to model a fuel-grade bioethanol production plant from unutilized rice straw as the feedstock. Process simulation-based inventory calculations along with the relevant literature-based inventory data are analyzed based on the ISO 14040/44 standard LCA methodology. Figure 01 illustrates the cradle-to-gate system boundary for the bioethanol production process using unutilized rice straw, considered in this study. Since unutilized rice straw is considered as the feedstock for this study, there is no inventory allocation from the paddy rice cultivation stage. Thus, the paddy rice cultivation stage can be excluded from the system boundary, and inventory data are evaluated starting from unutilized rice straw collection after paddy rice harvesting. Then the collected rice straw is dried, baled, and transported to the bioethanol processing plant.

Figure 01 (Page 25)

Cellulose and hemicellulose are the main compounds in rice straw which are convertible into sugars via fermentation. However, the presence of lignin in rice straw hinders cellulose and hemicellulose recovery. Therefore, a pretreatment operation is required for rice straw to remove lignin and recover cellulose and hemicellulose. In this study, the diluted acid pre-treatment technique with diluted sulfuric acid (1 w/v%) is considered as the pre-treatment method. The liquid phase containing cellulose and hemicellulose is then subjected to enzymatic hydrolysis to convert into glucose, xylose, and C_6/C_5 sugars. Thereafter, the solid phase containing lignin, ash, and other unconvertible residue is separated using a filter press. This solid residue can be used as a fuel to cogenerate process heat and electricity. The remaining acidic sugar solution is neutralized using $Ca(OH)_2$.

The neutralized sugar solution is undergone to simultaneous saccharification and fermentation (SSF) at the presence of yeast and $(NH_4)_2HPO_4$. After fermentation, a dilute ethanol solution (approximately 4.5 wt % of ethanol) is obtained and further purified using distillation methods. To produce fuel-grade bioethanol, anhydrous bioethanol (> 99.5 vol % purity of ethanol) is required. Therefore, the ethanol solution that is purified using initial distillation is further dehydrated to obtain fuel-grade bioethanol. Finally, the spent wash obtained from the distillation columns (in both initial distillation and dehydration units) is anaerobically digested (AD) to produce biogas. This biogas can be used as a fuel to cogenerate process heat and electricity for the same production process.

Energy consumption and generation in the modeled cradle-to-gate bioethanol production process is considered for the process net energy analysis and GHG assessment. Process simulations and inventory calculations are performed for the process based on the considerations as follows.

1. The life cycle considerations are reached by taking Sri Lanka as the location for this case study.
2. Dry rice straw in Sri Lanka has an average dry basis composition of cellulose: 30 wt %, hemicellulose: 3.9 wt %, lignin: 38 wt%, and others, including ash: 27.9 wt%. [28]
3. First, rice straw is separated after harvesting paddy rice. The average moisture content in rice straw is approximately 10 wt%.
4. The effects from infrastructure processes are negligible at the considered scale of this study [11].
5. Energy inputs for transportation and raw materials/chemicals manufacturing processes are derived from fossil energy sources.
6. Wastewater and spent-wash from bioethanol producing stages are treated by Upflow Anaerobic Sludge Blanket (UASB) reactors [29].
7. Recovered solid residues (after pre-treatment operation) and generated biogas are used to cogenerate process heat and electricity [27].
8. Surplus electricity generation is credited to the grid-mix electricity in Sri Lanka.
9. Gypsum coming out from neutralization and solid sludge generated from AD are used as fertilizers back in the paddy fields [23].
10. The opportunity loss of rice straw utilization for the most possible valorization option, i.e., application as manure, is compensated through:
    - Utilizing dried AD sludge and gypsum as fertilizers [30].
    - Leaving an uncut straw height at about 15 cm above the ground [7].
    - The environmental credit by avoiding the open burning of rice straw [23].

2.2 Inventory data calculations for cradle-to-gate bioethanol production process

For this study, bioethanol production of 1,000 L at 99.7 vol % purity is selected as the functional unit (FU). For a comprehensive analysis, the considered system boundary is sub-divided into five life cycle stages; 1. Feedstock drying and baling stage, 2. Feedstock and raw material transportation stage, 3. Feedstock pre-treatment stage, 4. Bioethanol conversion stage, and 5. Bioethanol dehydration stage.

Energy consumption and GHG emissions in each life cycle stage are calculated based on 1,000 L of bioethanol production at 99.7 vol% purity.

2.2.1 Feedstock baling and drying stage

According to process simulation results, 5.84 tonnes of dry rice straw are required to produce 1,000 L of 99.7 vol% bioethanol. The average diesel consumption of 1.2 L is required for the baling machine to bale one tonne of rice straw at a bale size of 0.2 m^3 (1 m×0.5 m×0.4 m) [25]. The Lower Heating Value (LHV) for diesel is 45.3 MJ/ L and density is 840 kg/m^3 [31, 32].

Baling and drying machine consumes 7.0 L of diesel/FU (5.88 kg of diesel/FU). GHG emission factors for diesel combustion in baling and drying machines are presented in Table S01 in the supplementary document. [23]

2.2.2 Feedstock and raw material transportation stage

The transportation stage consists of two main phases, i.e., transportation of feedstock from paddy fields to the bioethanol plant and transportation of required raw materials/chemicals from foreign countries to the bioethanol plant. Diesel trucks with a 10-tonnes capacity and a fuel economy of 4.5 L/km are considered for feedstock and raw material transportation [7]. The average round trip distance from the paddy fields to the bioethanol plant is taken as 100 km. Diesel volume for feedstock transportation is calculated using equation (01) [23].

$$Diesel\ volume\ (L) = \frac{distance\ (km) \times material\ amount\ (tonne)}{truck\ fuel\ economy\ \left(\frac{km}{L}\right) \times truck\ capacity\ (tonne)} \tag{01}$$

The volume of diesel required for feedstock transportation from paddy fields to the bioethanol plant is 12.97 L/FU. The energy consumption for transportation of chemicals from foreign countries to the local port is calculated from equation (02) [23].

$$E_{nautical} = 6{,}000\ (km) \times 0.08 \left(\frac{MJ}{tonne \cdot km}\right) \times material\ amount\ (tonne) \tag{02}$$

The average nautical distance is taken as 6,000 km where the average energy required to transport one tonne of material per one km via shipping is taken as 0.08 MJ/tonne.km [23]. The energy required to transport raw materials from the port to the bioethanol plant is calculated by multiplying the diesel volume consumption using equation (01) and the LHV value of diesel. Round trip distance from the port to the bioethanol plant is taken as 400 km.

Table S02 in the supplementary document summarizes the total energy consumption for transportation of each chemical used in the process.

Emission factors for each mode of transportation are given in Table S03 in the supplementary document [23]. Respective GHG emissions are calculated considering the transported weight, transportation distance, and emission factors. Required inventory data for the feedstock pre-treatment stage, bioethanol conversion stage, and bioethanol dehydration stage are obtained using the process simulation technique.

2.2.3 Process simulation for bioethanol processing stages

The bioethanol processing stages, such as the feedstock pre-treatment stage, the bioethanol conversion stage, and the bioethanol dehydration stage are simulated using the Aspen Plus process simulation software tool to obtain the required inventory data. The Aspen Plus property database is used to obtain the ethanol-water binary properties and ethanol-water-ethylene glycol ternary properties. There is an azeotrope in the ethanol and water binary mixture at 87 mol% (approximately 93 wt %) of ethanol at 1 atm. Due to this azeotrope, obtaining anhydrous bioethanol above 93 wt % purity cannot be performed using conventional distillation operations. Thus, dehydration techniques are used to break down the azeotrope. In this study, the extractive distillation technique using ethylene glycol as the extractive solvent is used for bioethanol dehydration. Extractive distillation is considered more energy efficient compared to other ethanol dehydration techniques [22].

The non-Random Two Liquid (NRTL) activity coefficient model is used as the thermodynamic property method for process simulations. The RadFrac rigorous distillation column model and the R-Stoic reactor model in the Aspen Plus model library are used to simulate distillation columns and reactors, respectively.

The energy requirements in the bioethanol production process are evaluated based on the obtained process simulation results. Energy requirement in each respective stage is compensated by in-situ heat and power cogeneration via combustion of biogas from the plant wastewater and lignin contained solid residue from feedstock pre-treatment.

Furthermore, GHG emissions in the respective stage are calculated considering the amount of energy and the source of energy utilized. In-situ biogenic energy does not involve carbon dioxide emissions due to the carbon-neutral rule, however, biogenic methane and nitrous oxide emissions are accounted for the GHG assessment. Emission factors for each biogenic energy source are represented in Table S04 in the supplementary document [23].

The contribution of biogas and lignin contained solid residue for heat and electricity generation were considered, separately. Thus, percentage contributions for heat generation from biogas and lignin contained solid residue are 2.53% and 97.47%, respectively. As well, percentage contributions for electricity generation are distributed between biogas and lignin contained solid residue at 8.14 % and 91.86 %, respectively.

(a) Feedstock pre-treatment stage

This stage consists of processing operations, such as crushing, pre-treatment, enzyme hydrolysis, and neutralization before the fermentation unit. A knife mill with a power consumption of 5 kWh and a capacity of 200 kg/hr is used to crush unutilized rice straw (at < 10 mm particle size). The average energy required to crush one tonne of rice straw is 99.0 MJ [7].

The reactions and process conditions in pre-treatment, hydrolysis and neutralization operations are given in Table S05 in the supplementary document.

(b) Bioethanol conversion stage

This stage comprises the SSF unit and distillation unit for initial bioethanol conversion and purification. Reactions and process conditions are given in Table S06 in the supplementary document.

(c) Bioethanol dehydration stage

Further purification of bioethanol to reach 99.7 vol % purity using the extractive distillation unit that consists of a dehydration column and ethylene glycol recovery column are considered in this processing stage. Process conditions for the bioethanol dehydration stage are given in Table S07 in the supplementary document.

(d) Chemicals and raw materials manufacturing for bioethanol processing stages

Table S08 in the supplementary document summarizes the average energy consumption for required chemicals and raw materials manufacturing. The energy content of direct steam used in the pre-treatment unit is calculated using steam properties. Enthalpy of saturated steam at 3.5 bar is 2.732 MJ/kg. The process consumes 2,237.23 kg of steam per FU. Thus, the energy content of direct steam used is 6,112.11 MJ/FU.

Table S09 in the supplementary document represents emission factors and GHG emissions of each chemical utilized in the bioethanol production phase.

(e) Calculations for process energy supply

The lignin contained solid residue and biogas generated from AD are the main two fuels used to cogenerate process steam and electricity. The total quantity of lignin contained solid residue is 4,055.61 kg/FU. The amount of biogas (65% CH_4) production is determined using equation (03) [33].

$$Generated\ Methane\ Amount(kg) = wastewater\ volume\ \times COD \times B_0 \times MCF \qquad (03)$$

where, B_0 is the maximum methane producing capacity (i.e., 25 kg-CH_4/kg-COD) and MCF is the Methane Correction Factor (i.e., 0.8) [33]. Wastewater volume generated in the process is 18.25 m^3/FU. COD value was calculated considering the composition of wastewater. Table S10 in the supplementary document represents the wastewater composition.

The calculated COD value of wastewater is 14.41 kg/m^3. Thus, the amount of CH_4 generated is 73.64 m^3/FU, and generated biogas amount is 113.29 m^3/FU.

Combined Heat and Power (CHP) units with a steam turbine and a gas turbine are used to cogenerate process heat and electricity using lignin contained solid residue and biogas as fuels, respectively. Fuel heating values, efficiencies of CHP units, and cogenerated heat and power amounts are summarized in Table S11 in the supplementary document.

(f) Creditable GHG emissions

The bioethanol production itself utilizes green energy throughout the process and there is a surplus electricity and steam generation within the plant. This surplus electricity is credited to Sri Lankan national electricity grid mix. Accordingly, the corresponding emissions can be credited, including the GHG credit. Table S12 in the supplementary document represents the emission factors for the Sri Lankan electricity grid mix [34].

2.3 Methodology for net energy analysis

Net energy inputs and net energy outputs are calculated to evaluate net energy indicators for the assessment of energy sustainability of the considered bioethanol production life cycle. Four energy indicators are calculated under the net energy analysis in this study, given in equations (04), (05), (06), and (07).

$$Net\ Energy\ Value\ (NEV) = total\ net\ energy\ outputs - total\ net\ energy\ inputs \qquad (04)$$

$$Net\ Energy\ Ratio\ (NER) = \frac{total\ net\ energy\ outputs}{total\ net\ energy\ inputs} \qquad (05)$$

$$Net\ Renewable\ Energy\ Value\ (NRnEV) = total\ net\ bioenergy\ output - total\ net\ fossil\ energy\ input \tag{06}$$

$$Renewability\ (Rn) = \frac{total\ net\ bioenergy\ outputs}{total\ net\ fossil\ energy\ inputs} \tag{07}$$

2.4 Methodology for Global Warming Potential (GWP)

The GWP is a measurement of the total GHG emissions from an activity, both directly and indirectly, or accumulated over the considered life cycle. The overall GWP value is calculated using equation 08 considering individual GHG emission amounts and their characterization factors for the global warming potential.

$$GWP = \sum GWP_i \times m_i \tag{08}$$

GWP_i = Global Warming Potential value of substance i

m_i = amount of substance i

The GWP_i values for individual GHGs, such as CO_2, CH_4, and N_2O are given in Table S13 in the supplementary document.

3. Results and discussion

3.1 Process simulation results

Figure 02 illustrates the process simulation flowsheet, including detailed material and energy flows. According to simulation results, the bioethanol yield (at 99.7 vol % purity) from unutilized rice straw in the scaled-up process is 171.34 L /tonne of rice straw (dry basis). The scaled-up bioethanol plant was simulated in the Aspen Plus process simulation software to obtain 1,000 L of bioethanol at 99.7 vol % purity. According to the simulation results, 5.84 tonnes of unutilized rice straw (dry basis) is required to produce 1,000 L of bioethanol at 99.7 vol % purity (FU).

Figure 02 (Page 26)

Table 01 lists the process simulation-based results for process energy consumption in each plant equipment in the scaled-up plant, including the rice straw crusher for pre-processing. The total process energy consumption by all plant equipment is 24,314.8 MJ/FU where the total steam consumption is 16,378.4 MJ/FU and the total electricity consumption is 7,936.3 MJ/FU.

Table 01 (Page 30)

Table 02 shows bioethanol yield and total process energy input, reported in other published studies in comparison to the same parameters in this study. Accordingly, the simulated process energy input in this study is comparable with that of bioethanol production plants, reported in the published literature. However, the bioethanol yield in this study is lower compared to other studies. The plausible reason is the lower cellulose and hemicellulose content in Sri Lankan rice straw that was considered in this case study. Therefore, process simulations performed in this study can be validated for the retrieval of life cycle inventory data.

Table 02 (Page 31)

Energy inputs for each life cycle stage in the considered system boundary are calculated considering the process energy consumption and energy uptake for manufacturing and transportation of raw materials/chemicals. The total heat consumption, total electricity consumption, and percentage energy consumption for each life cycle stage are summarized in Table 03.

Table 03 (Page 32)

Figure 03 indicates the graphical representation of stage-wise energy consumption. The feedstock pre-treatment stage is responsible for 61.1 % of the total cradle-to-gate energy input, which is the highest energy up-taking stage. The bioethanol conversion stage corresponds to the second highest energy consumption (25.7 % of the total).

The bioethanol dehydration stage consumes only about 8.3 % of the total energy consumption, which implies that converting hydrous bioethanol to fuel-grade anhydrous bioethanol is not highly energy-intensive compared to other upstream bioethanol conversion operations. Thus, upgrading an existing bioethanol plant to obtain fuel-grade bioethanol via extractive distillation is feasible without incurring high energy demand for the upgraded section of the bioethanol plant.

Figure 03 (Page 27)

3.2 Life cycle net energy analysis

Table 04 indicates the life cycle net energy balance, including the calculated net energy indicators for the cradle-to-gate bioethanol production from unutilized rice straw in Sri Lanka. The total net energy input is 38,938.9 MJ/FU, which contains the total net fossil fuel energy input of 8,512.0 MJ/FU. The total net energy output is 46,742.8 MJ/FU, which is entirely a bioenergy output (surplus process energy + energy content in bioethanol). Calculation results for NEV and NRnEV are 7,804.0 MJ/FU and 38,230.9 MJ/FU, respectively, as well as NER and Rn for the process, are 1.20 and 5.49, respectively.

Table 04 (Page 33)

Figure 04 illustrates the resulting net energy indicators in graphical form. A net bioenergy surplus is observed in the process when comparing the process energy consumption with generation. Thus, NEV and NRnEV for the life cycle of bioethanol production from rice straw in this study are positive values where the total net energy output is greater than the total net energy input, including fossil energy inputs. This implies that the considered cradle-to-gate bioethanol production process in this study is self-sufficient in terms of energy.

Figure 04 (Page 28)

Table 05 lists the net energy indicators and GWP values of relevant published LCA studies on bioethanol production using rice straw, comparable to this study. According to Table 05, some of the published LCA studies for bioethanol production using rice straw that included inventory allocation from the rice cultivation stage have reported negative values for NEV and NRnEV indicators as well as NER and Rn values lesser than 1. Even though positive values have been reported, the net energy indicator values in other published studies are lower compared to that of this study. Hence, the introduced life cycle consideration for unutilized rice straw in this study (zero inventory allocation from cultivation stage) has affected to turn the life cycle of bioethanol production from rice straw more sustainable and renewable. Therefore, the net energy analysis results in this study indicate that the life cycle consideration of zero inventory allocation for the rice cultivation stage is a determinant factor in future LCA studies for the cases/scenarios of unutilized rice straw valorization through bioethanol production.

Table 05 (Page 34)

3.3 Life cycle GHG assessment

Table 06 presents the GHG emissions in respective life cycle stages and the creditable GHG amounts. According to the calculation results shown in Table 06, the net GWP value for 1,000 L of bioethanol at 99.7 vol % purity is 584.76 kg CO_2 eq./FU. This GWP value is significantly lower compared to various published LCA studies on bioethanol production from rice straw. The major reason for this reduction of GWP value is the cancellation of GHG emissions from the rice cultivation stage implied by the life cycle consideration for unutilized rice straw. In addition, the scaled-up bioethanol production plant which was simulated with improved energy efficiency and waste recovery methods, also contributes to a lower total GWP value, compared to that of other bioethanol plants reported in the literature. Thus, the result findings and the methodologies in this study contribute to designing new scaled-up process plants for more environmentally benign bioethanol production using unutilized rice straw as the feedstock.

Table 06 (Page 35)

Figure 05 depicts the graphical interpretation of GHG emissions in each life cycle stage. The highest amount of GHG is emitted in the feedstock pre-treatment stage that corresponds to 803.17 kg CO_2 eq. / FU. The bioethanol dehydration stage has the least significant GHG emissions (3.91 kg CO_2 eq./FU) which interprets its very low influence on environmental impacts compared to that of other life cycle stages in the bioethanol production life cycle.

However, there can be a policy-wise reluctance and social barriers in developing countries like Sri Lanka assuming that a high energy intake and increased GHG emissions would be there for fuel-grade bioethanol production. This dilemma would restrain the valorization of unutilized rice straw via bioethanol production and upgrading of existing bioethanol production plants up to fuel-grade bioethanol purity. Nevertheless, this study provides useful findings to promote policy decision-making for upgrading any existing plant and establishment of new plants for fuel-grade bioethanol production from unutilized rice straw as the feedstock, with more environmental sustainability.

Figure 05 (Page 29)

3.4 Sensitivity analysis

For interpretation of the sensitivity of the results from life cycle net energy analysis and GHG assessment in this study, a sensitivity analysis is performed for a ± 5% range of variation of two key process parameters, i.e., bioethanol yield (L/tonne of rice straw) and process energy consumption (MJ/FU). The sensitivity of the impact indicators: NER, Rn, and GWP are observed concerning the key process parameter variations. Table 07 indicates the results from the sensitivity analysis. The variations of the two key process parameters within a range of ±5%, do not inhibit the energy sustainability of the process and affect the GWP values at tolerable levels.

Table 07 (Page 36)

The process energy consumption holds the highest sensitivity for all three impact indicators resulting in wide ranges for the respective variation in process energy consumption. Among the impact indicators, GWP has the highest sensitivity towards the variation of process energy consumption. Decrease in process energy consumption from 24,314.77 MJ/FU to 23,099.03 MJ/FU improves the NER from 1.20 to 1.27, Rn from 5.49 to 5.63, and GWP from 584.76 kg CO_2 eq./FU to 524.25 kg CO_2 eq. /FU. Thus, increasing the process energy efficiency further improves the energy sustainability and reduces the GWP value of the bioethanol production process from unutilized rice straw. For the variation of bioethanol yield, NER shows the highest variation and a negligible variation in GWP value. However, higher process yields improve the energy sustainability of the process and lower the GWP as well. Therefore, collaborative improvements in both bioethanol yield and process energy efficiencies will secure the energy sustainability of fuel-grade bioethanol production from unutilized rice straw as the feedstock while reducing the climate change impact with lower GWP values.

3.5 Economic estimation and GHG credits for system boundary expansion

Fuel-grade bioethanol can be blended with gasoline at different proportions that produce various gasohol types (E3, E10, E20, E85, E87, E100, etc.). Gasohol from E3 to E85 can be used in vehicles without any engine modification. Hence, a system boundary expansion of E10 gasohol production and its end use is considered in this study in terms of economic estimation and GHG credits. If E10 gasohol is produced, 10% gasoline imported to Sri Lanka can be substituted from fuel-grade bioethanol, produced locally using unutilized rice straw that corresponds to 22,400 m^3 of bioethanol per year.

Table 08 summarizes the economic estimation and GHG assessment results for the system boundary expansion. According to economic estimation results, an annual net import cost of about USD 13.62 million can be saved by producing E10 gasohol within the country. GHG assessment results considering the system boundary expansion

(Utilization of E10 gasohol by substituting 10% of gasoline combusted in vehicles) shows a GHG credit of more than 40,837 tonnes of CO_2 eq. per year. In addition, this system boundary expansion accounts for further GHG credits considering the avoidance of open burning of unutilized rice straw without compensating it for the opportunity loss of rice straw as manure. Avoidance of open burning of rice straw credits a GHG amount of 92 kg of CO_2 eq. per tonne of rice straw that corresponds to 12,027 kg of CO_2 eq, per year. Thus, the net GHG credit from both E10 gasohol substitution and avoidance of open burning of rice straw is approximately 39,766 tonnes of CO_2 eq. per year. Therefore, the system boundary expansion from cradle-to-gate to cradle-to-grave makes the bioethanol production lifecycle using unutilized rice straw entirely carbon-negative, which is an attractive opportunity for policy decision making in developing countries like Sri Lanka.

Table 08 (Page 37)

Conclusion

This study concludes that valorization of unutilized rice straw to produce fuel-grade bioethanol reaches higher net energy gain and renewability along with a lower GWP, compared to reported real plants in the published literature. Hence, consideration of zero inventory allocation from the paddy rice cultivation stage for unutilized rice straw as a bioethanol feedstock will be a determinant factor for future studies and the establishment of new bioethanol plants. Fuel-grade bioethanol production from unutilized rice straw contributes to replacing gasoline imports and consumption for a country like Sri Lanka with an agricultural economy that cultivates paddy rice as a major crop. The associated economic benefits with foreign currency savings are significant as revealed in this study. Further, the use of unutilized rice straw while avoiding open burning along with gasohol blending provides a net GHG credit. In addition, the life cycle stage-wise net energy analysis and GHG assessment reveal that the bioethanol dehydration stage consumes a low energy amount and is responsible for low GHG emissions, compared to that of other life cycle stages. Thus, the findings from this study with a simulated process plant would support the decision-making for upgrading existing bioethanol plants and establishment of new fuel-grade bioethanol plants through the valorization of unutilized rice straw at commercial-scale in the future.

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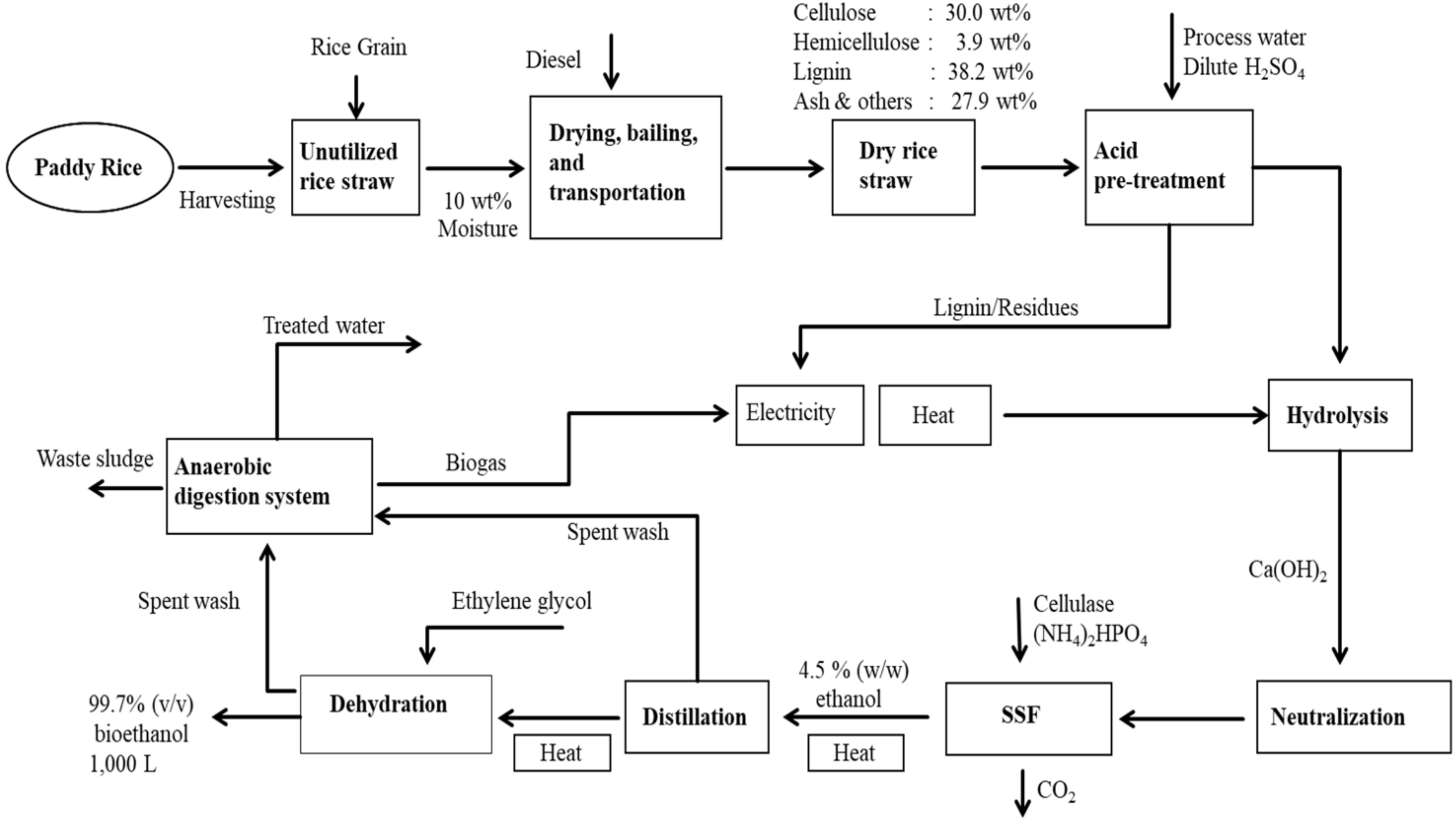

Figure 01: Cradle-to-gate system boundary of bioethanol production process from unutilized rice straw

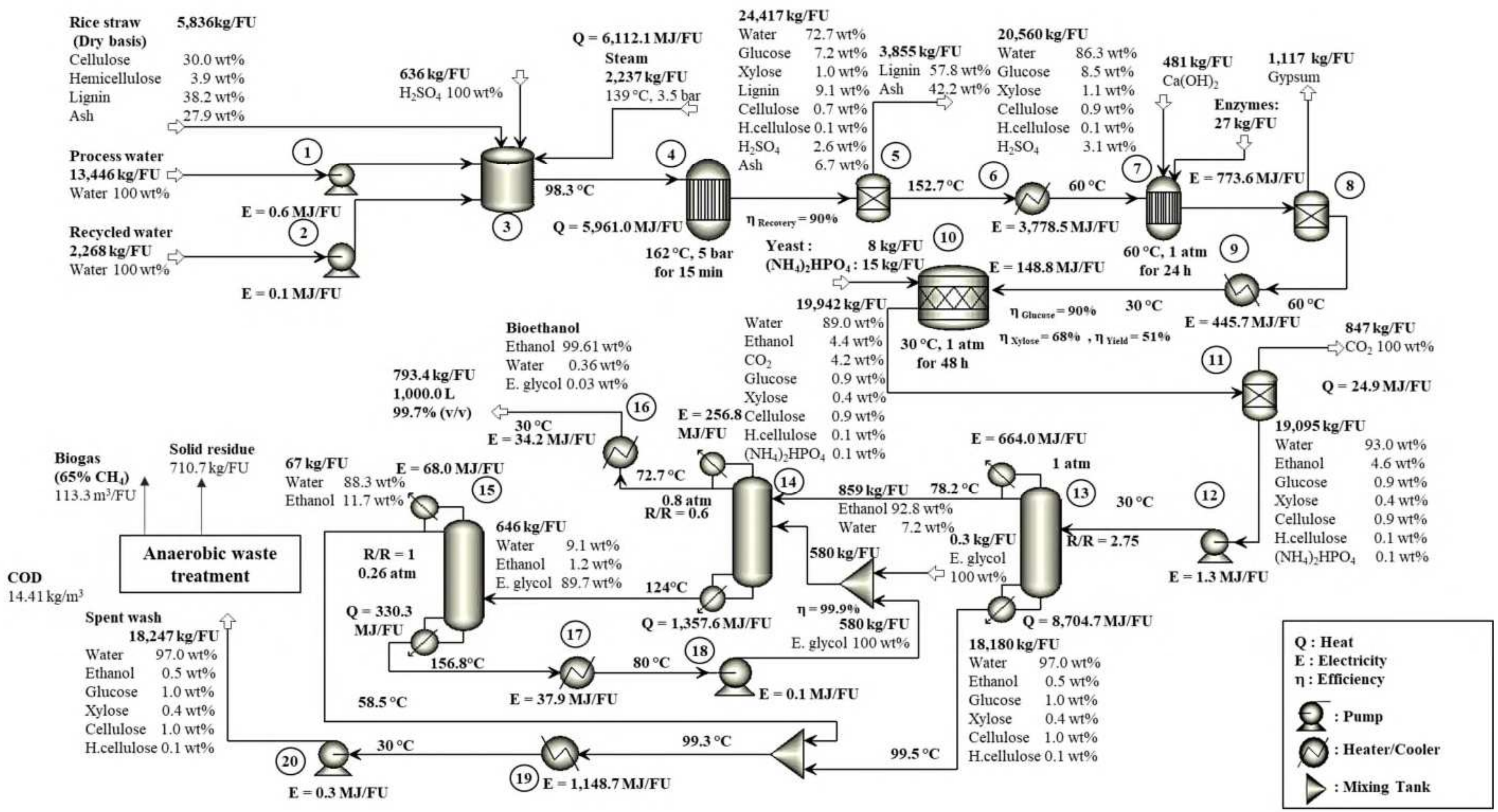

Figure 02: Process simulation flowsheet for scaled-up bioethanol production plant from unutilized rice straw. Where, **1:** Pump no.1, **2:** Pump no. 2, **3:** Mixing tank, **4:** Pre-treatment unit, **5:** Filter no. 1, **6:** Cooler no. 1, **7:** Detoxifying unit, **8:** Filter no. 2, **9:** Cooler no. 2, **10:** SSF, **11:** Scrubber, **12:** Pump no. 3, **13:** Column no. 1, **14:** Column no. 2, **15:** Column no. 3, **16:** Cooler no. 3, **17:** Cooler no. 4, **18:** Pump no. 4, **19:** Cooler no. 5, **20:** Pump 5

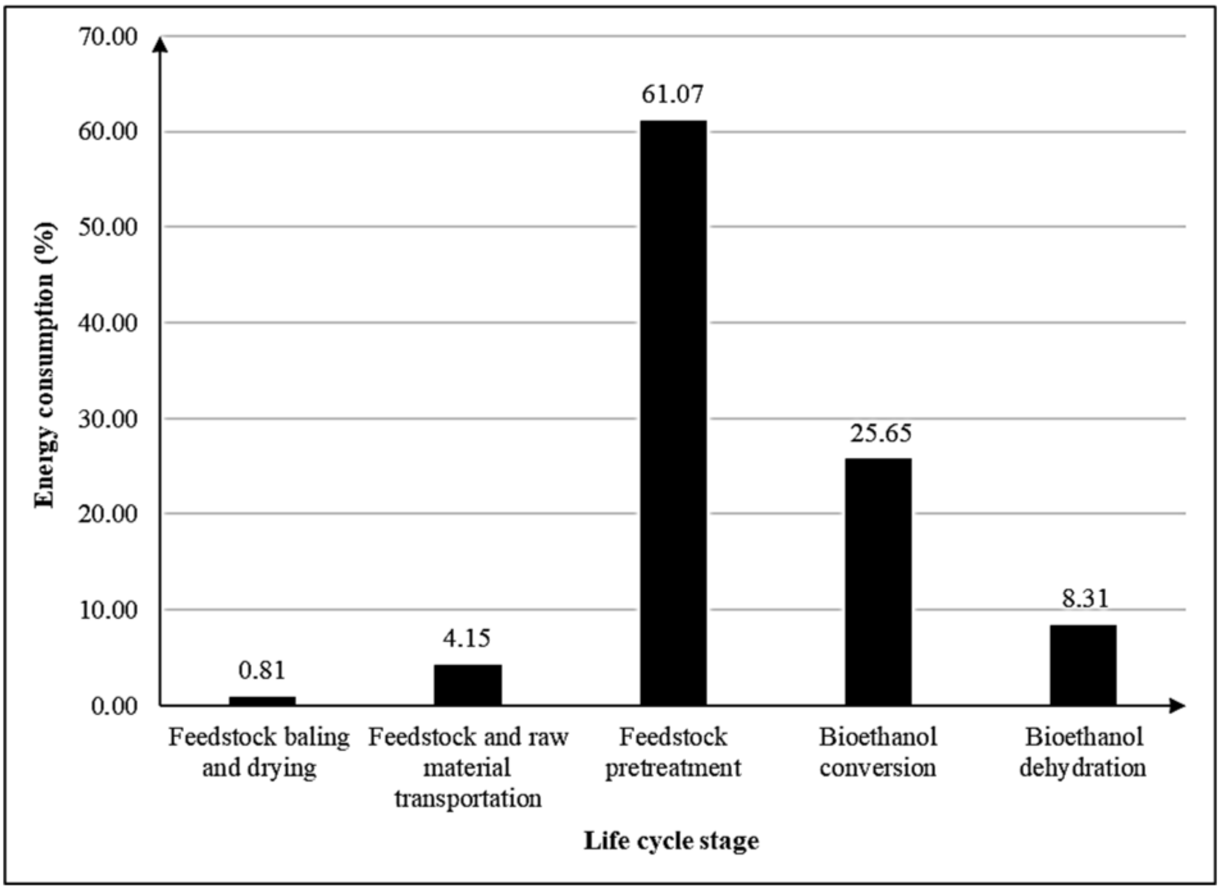

Figure 03: Life cycle stage-wise energy consumption in cradle-to-gate system boundary

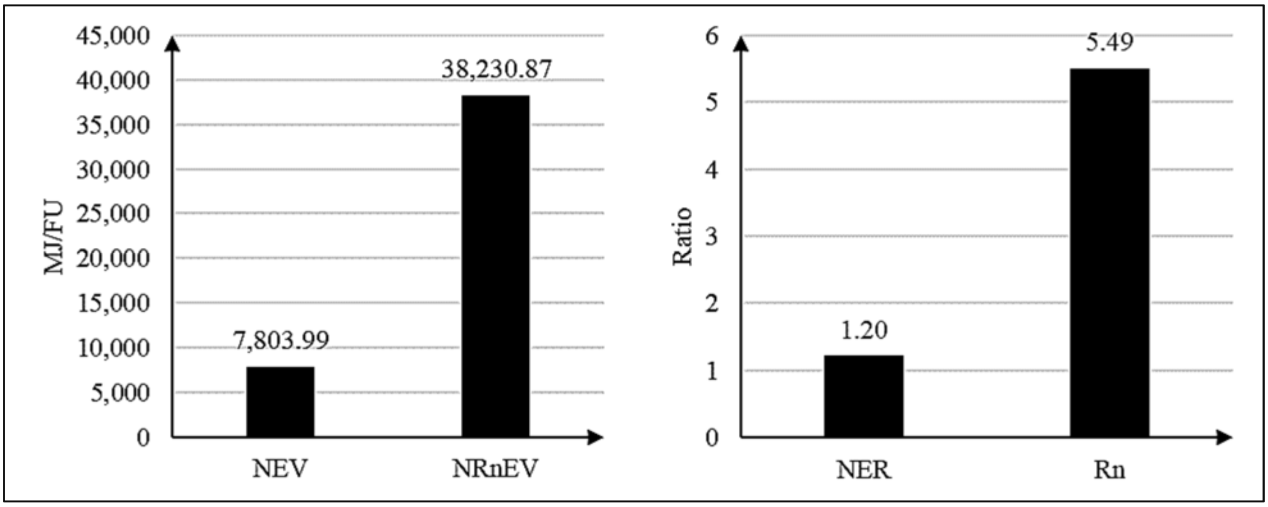

Figure 04: Net energy indicator results

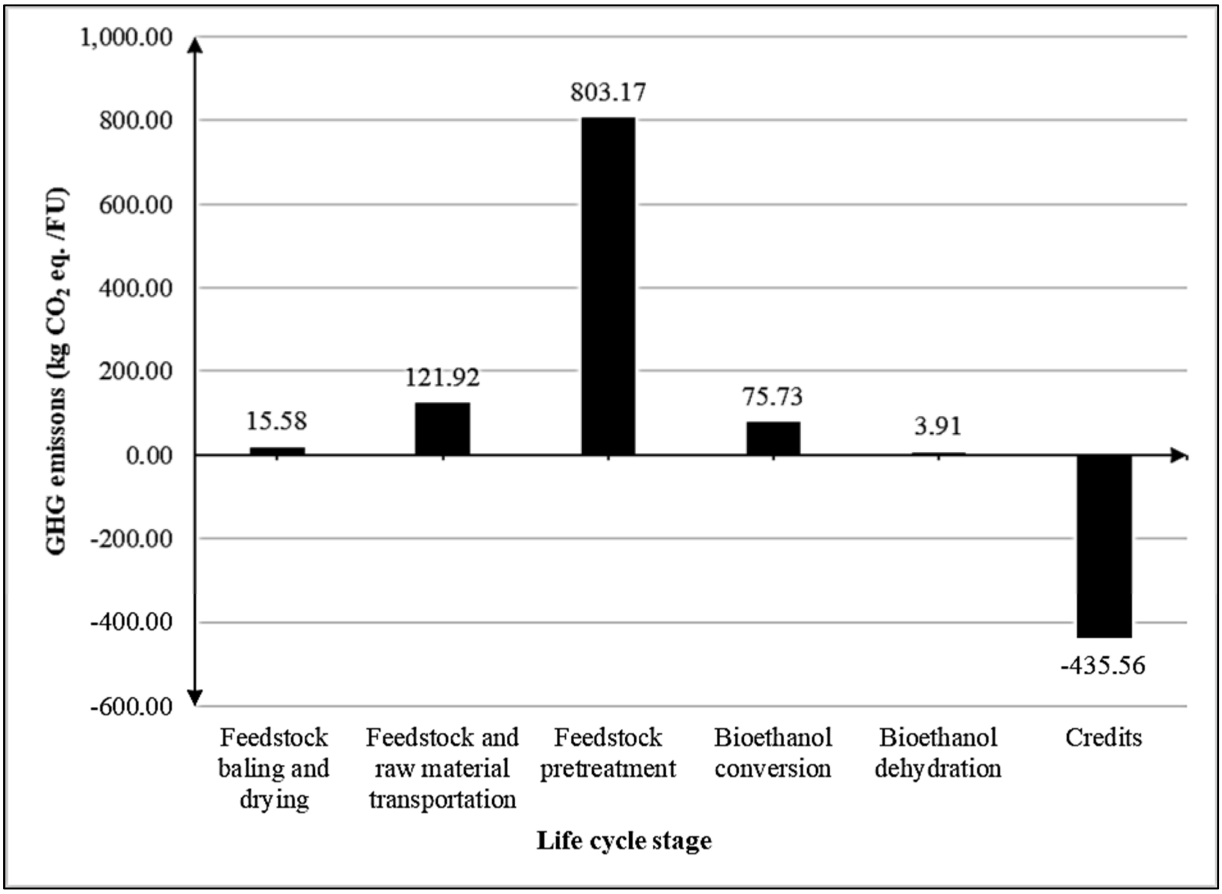

Figure 05: Life cycle stage-wise GWP for bioethanol production from unutilized rice straw

Table 01: Energy consumption results for individual plant equipment (basis: 1,000 L of bioethanol at 99.7 vol% purity)

No	**Equipment**	**Energy consumption (MJ/FU)**		
		Steam	**Power**	**Total**
1	Crusher	-	577.8	577.8
2	Pump no. 1	-	0.6	0.6
3	Pump no. 2	-	0.1	0.1
4	Mixing tank	-	0.05	0.05
5	Pretreatment unit	5,961.0	-	5,961.0
6	Filter no. 1	-	0.03	0.03
7	Cooler no. 1	-	3,778.5	3,778.5
8	Detoxifying unit	-	773.6	773.6
9	Filter no. 2	-	0.02	0.02
10	Cooler no. 2	-	445.7	445.7
11	SSF	-	148.8	148.8
12	Scrubber	24.9	-	24.9
13	Pump no. 3	-	1.4	1.4
14	Column no. 1	8,704.7	664.0	9,368.6
15	Column no. 2	1,357.6	256.8	1,614.4
16	Column no. 3	330.3	67.9	398.3
17	Cooler no. 3	-	34.2	34.2
18	Cooler no. 4	-	37.9	37.9
19	Pump no. 4	-	0.1	0.1
20	Cooler no. 5	-	1,148.7	1,148.7
21	Pump no. 5	-	0.3	0.3
22	**Total**	**16,378.4**	**7,936.4**	**24,314.8**

Table 02: Literature-based comparison for process parameters

Country	**Process ethanol yield (L/tonne of rice straw)**	**Process energy consumption (MJ/FU)**			**References**
		Heat	**Electricity**	**Total**	
Thailand	260.0	17,467.0	5,703.0	23,170.0	[23]
India	239.0	16,345.5	2,048.5	18,394.0	[7]
Japan	250.0	-	-	11,560.0	[35]
Japan	373.0	-	-	22,890.0	[26]
Thailand	260.0	-	-	28,734.0	[25]
Sri Lanka	171.3	16,378.4	7,936.4	24,314.8	This Study

Table 03: Energy consumption results for process stages (basis: 1,000 L of bioethanol at 99.7 vol% purity)

Stage	Energy consumption (MJ/FU)			
	Fossil	**Heat**	**Electricity**	**Total per stage**
Feedstock baling and drying	317.28	-	-	317.28
Feedstock and raw material transportation	1,617.76	-	-	1,617.76
Feedstock Pretreatment				23,780.06
1. Chemicals / raw materials	6,130.67	-	-	
2. Direct steam	-	6,112.11	-	
3. Process	-	5,960.95	5,576.33	
Bioethanol conversion				9,986.04
1. Chemicals / raw materials	442.36	-	-	
2. Process	-	8,729.54	814.13	
Bioethanol dehydration				3,237.72
1. Chemicals / raw materials	3.90	-	-	
2. Process energy	-	1,687.95	1,545.87	
Total	**8,511.97**	**22,490.55**	**7,936.33**	**38,938.85**

Table 04: Net energy balance for cradle-to-gate bioethanol process from unutilized rice straw

Description	Energy input/output (MJ/FU)		
	Heat	**Electricity**	**Fossil**
Feedstock baling and drying stage	-	-	317.3
Feedstock and raw material transportation stage			
Feedstock transportation	-	-	587.6
Raw Materials/Chemicals transportation			1,030.2
Feedstock pretreatment stage			
Raw material manufacturing	6,112.1		6,130.7
Direct steam	-	-	-
Energy consumption	5,961.0	5,576.3	-
Bioethanol Conversion stage			
Raw material manufacturing	-	-	442.4
Energy consumption	8,729.5	814.1	-
Bioethanol Dehydration stage			
Raw material manufacturing	-	-	3.9
Energy consumption	1,687.9	1,545.9	-
Total energy consumption	22,490.6	7,936.4	8,512.0
Energy production (Lignin residue + Biogas)	45,064.2	10,905.5	-
Surplus energy	22,573.6	2,969.2	-
Energy content in 1,000 L of bioethanol at 99.7 vol % purity		21,200.0	
Total net energy inputs		38,938.9	
Total net fossil energy inputs		8,512.0	
Total net energy outputs		46,742.8	
Total net bioenergy outputs		46,742.8	
NEV		7,804.0	
NRnEV		38,230.9	
NER*		1.20	
Rn*		5.49	

* Dimensionless parameters

Table 05: Results comparison of published LCA studies in existing literature

Country	NEV MJ/FU	NRnEV MJ/FU	NER	Rn	GHG emissions (kg CO_2 eq./FU)	Reference
Thailand	(4,331.00)	18,840.00	0.85	3.92	1,502.00	[23]
Europe	(1,252.60)	-	0.20	-	-	[36]
Japan	1,648.00	-	1.17	-	-	[26]
Japan	-	-	-	-	1,145.00	[35]
India	6,978.00	14,900.00	1.36	2.30	1,222.00	[7]
Sri Lanka	7,803.99	38,230.87	1.20	5.94	584.76	This Study

Table 06: Life cycle stage-wise GHG emission results (basis: 1,000 L of bioethanol at 99.7 vol% purity (FU))

Stage/phase	**GHG emissions (kg CO_2 eq./FU)**				
	CO_2	**CH_4**	**N_2O**	**Total GHG**	**Grand total**
01. Feedstock bailing and drying	15.53	0.01	0.04	15.58	15.58
02. Feedstock and raw material transportation					
Feedstock transportation	43.19	0.01	0.28	43.48	121.92
Nautical transportation of raw materials	43.42	0.04	0.42	43.87	
Inland transportation of raw materials	34.55	0.001	0.02	34.57	
03. Feedstock Pretreatment					
Biogas combustion	-	6.66	0.29	6.95	803.17
Lignin combustion	-	0.85	2.61	3.45	
Direct steam	-	2.17	1.49	3.66	
Chemicals / raw materials	789.11	-	-	789.11	
04. Bioethanol conversion stage					
Biogas combustion	-	3.16	0.14	3.30	75.73
Lignin combustion	-	0.72	2.21	2.92	
Chemicals / raw materials	69.51	-	-	69.51	
05. Bioethanol dehydration					
Biogas combustion	-	1.86	0.08	1.94	3.91
Lignin combustion	-	0.24	0.73	0.97	
Chemicals / raw materials	1.00	-	-	1.00	
06. Credits					
Surplus energy	(452.71)	10.87	6.28	(435.56)	(435.56)
Total GHG emissions					**584.76**

Table 07: Sensitivity analysis results

Description	Sensitivity parameter	
	Bioethanol yield (L/tonnes of rice straw)	Process energy (MJ/FU)
Parameter value in this study	171.34	24,314.77
Decreased / increased range*	162.78 - 179.91	23,099.03 – 25,530.51
NER	1.18 – 1.22	1.27 – 1.13
Rn	5.43 – 5.55	5.63 – 5.35
GWP	586.60 – 583.09	524.25 – 636.82

* ±5% variation in sensitivity parameters

Table 08: Economic estimation results for E10 gasohol from rice straw bioethanol

Description	Unit	Value
Gasoline (Octane 95) imports* [37]	m^3/year	224,000
Gasoline importing cost per year [37]	USD/m^3	607.21
Gasoline substitution from E10 gasohol per year	m^3/year	22,400
Net Import Cost Saving	Millions USD/year	13.62
GHG credit for substituting gasoline [7]	kg of CO_2 eq./ FU	1,823.08
GHG credit by substituting from E10 gasohol	tonnes of CO_2 eq./year	40,836.92
GHG credit for avoiding open burning of rice straw [7]	kg of CO_2 eq./tonnes of rice straw	92
GHG credit by substituting from E10 gasohol + avoiding open burning of rice straw	**tonnes of CO_2 eq./year**	**52,864.27**
Total GHG emission for bioethanol production	**tonnes of CO_2 eq./year**	**13,098.56**
Net GHG credit	**tonnes of CO_2 eq./year**	**39,765.71**

*Available recent data as per the year 2017

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