Sustainability assessment of levulinic acid and succinic acid production from empty fruit bunch

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Abstract. Empty fruit bunch (EFB) generated as waste in plantation mill activities in Malaysia is a potential biomass feedstock of biorefinery for fuels and chemicals production. Levulinic acid and succinic acid, two out of 12 chemical building blocks identified by Department of Energy (DOE) to be used in synthesis of high-value materials, can be produced from biochemical conversion of the EFB. This paper evaluates sustainability assessment of EFB to levulinic acid and succinic acid. The assessments include net present value (NPV), global warming potential (GWP), and Hazard identification and ranking (HIRA) to cater for economic, environment, and safety performances, respectively. The results show that the levulinic acid production is more economically attractive than succinic acid production. The environmental impact quantification reveals that the levulinic acid has lower GWP score of 6.3 kgCO2-eq/kg levulinic acid than succinic acid with 11.2 kgCO2-eq/kg succinic acid. Meanwhile, the succinic acid production is inherently safer than levulinic acid production due to its less severe operating conditions.

1. Introduction

Nowadays, the current fossil fuels encounter the sustainability issues of depletion and environmental impact. The substitution of fossil fuels with renewable source utilization will resolve the problem of energy scarcity and greenhouse gas (GHG) emissions [1]. Malaysia poses plenty of biomass residue especially empty fruit bunch (EFB) from its milling and plantation activity which created a major disposal problem [2]. In fact, for every ton of palm oil generated 22% of empty fruit bunches (EFBs) and characteristically, EFB consists of cellulose (24-65%), hemicellulose (21-34%), and lignin (14-31%). Thus, EFB has potential as source of fermentable sugars to produce value-added chemicals because of high content cellulose and hemicellulose [3].

The Department of Energy (DOE) has categorized 12 chemical building blocks that can be obtained from biomass as a platform chemical [4]. Among those chemicals, levulinic acid (LA) and succinic acid (SA) are favorable organic intermediate to synthesize wide range of chemicals [5]. LA can be derived from carbohydrates of biomass and it is produced by way of hydrolysis of cellulose into glucose monomers then followed by dehydration of glucose into HMF (Hydroxymethylfurfural) which transform it into LA [6]. It is used in many applications for instance pharmaceuticals, plasticizers, fragrances, and cosmetics. Meanwhile, SA is produce through the hydrogenation of petroleum-based maleic anhydride with the severe operating conditions and costly catalyst [7]. However, the biochemical fermentation process has introduced to produce SA with moderate operating condition and lead to cost-competitive. SA can be used to produced food and pharmaceutical products, surfactants, and detergents, green solvents and biodegradable plastics [8].

The previous studies of sustainability assessment both levulinic acid and succinic acid focused solely on economic performance and environmental impact. The study of [9] investigated a sustainability aspect in terms of environment, energy consumption, and economic for two scenarios of levulinic acid biorefinery value chain. [10] presented the sustainability of succinic acid via
fermentative synthesis and the petrochemical route by considering total cost production and energy efficiency. Unfortunately, a safety aspect has not been the consideration in the biorefinery assessment for both processes. However, the challenges exist to hinder the sustainable process such as the complexity of the process, critical operating conditions, toxic and flammable materials handling, and treatment hazardous materials [11]. The activity for understanding the hazard at initial design can be invaluable to eliminate and reduce the hazard in order to build an inherently safer design (ISD) of a chemical process [12]. Currently, the economic and environmental criteria are still majorly used for making the decision in process design development of the integrated biorefinery. Meanwhile, the failure of hazard identification at an early stage in process design leads to the chemical accidents. Therefore, the chemical plant is expected to pose less hazardous potential to make it more safe [13]. Therefore, the aim of this study is to perform a sustainability assessment of both levulinic acid and succinic acid production from EFB taking into account economic, environment, and safety at an early design stage. To quantify the economic feasibility, the net present value (NPV) will be calculated. Furthermore, the life cycle assessment (LCA) and inherently safer methods are used to evaluate the global warming potential (GWP) and hazard identification and ranking (HIRA), respectively. In addition, the comparison between two processes in regard to sustainability assessment will be conducted.

2. Methodology

This paper will simulate and evaluate the sustainability assessment for both the production of levulinic acid and succinic acid from empty fruit bunch (EFB). The process design and simulation for both processes are carried out using Aspen Plus V10 and the process design associated to operation conditions, equipment consideration, conversion, pre-treatment method, and assumption are primarily based on [14-16]. The assessment of the sustainability in terms of economic, environmental, and safety aspect for both processes are conducted to calculate the net present value (NPV), global warming potential (GWP), and hazard and identification ranking (HIRA), respectively.

2.1. Process design

The simulation of the processes was performed using Aspen Plus V.10. An unavailable database in Aspen Plus was predicted by the properties of some components involved in [14]. In this work, the nonrandom two-liquid (NRTL) was selected as the thermodynamic package since it is good for non-ideal systems and wide boiling point.

2.1.1. Pretreatment. Figure 1a shows a simple block diagram that is created to represent the acid hydrolysis pretreatment process. This process route was based on a benchmark model from [32]. In the first part, EFB was separated into cellulose, hemicellulose, and lignin through acid hydrolysis with sulfuric acid as a catalyst, at elevated temperatures (158 – 200 oC). Upon treatment, the biomass was treated to remove lignin (delignification) leaving cellulose and hemicellulose fractions were converted into soluble-low-molecular weight components. Pre-treatment process serves to increase the accessibility of cellulose. An on-site enzyme production process was required to produce cellulase for conversion of cellulose in EFB into hexose sugars via the saccharification process at 90% conversion. The hexose sugars produced in these processes were then sent to conversion process to produce the levulinic acid and succinic acid.
Acid hydrolysis (xylan into xylose using H₂SO₄)

Saccharification (cellulose into glucose)

Enzyme production

To upgrading process

EFB

T = (158-200)°C
P = (5.6-5.7) atm
τ = (5-30) mins

T = 48°C
P=1 atm
τ = 3.5 days

Nutrients, SO₂, Glucose

Enzyme production

T = (158-130)°C
P = (5.6-5.7) atm
τ = (5-30) mins

Separation (lignin from hydrolysates)

Dehydration (dextrose, galactose and mannose)

Hydration of HMF

Burning of lignin solids

Separation of lignin from hydrolysates

Energy as fuel to process

Heavies

Energy as fuel to process

Water (may not be required)

Recovery and purification of LA

[lit][SA]

Figure 1. The process flow diagram: (a) pretreatment process, (b) levulinic acid production, and (c) succinic acid production.

2.1.2. Levulinic acid production. Figure 1b shows a simple block diagram that was created to represent the process to produce levulinic acid. This process route was based on experimental data from [15]. First, the hexose sugars derived from pretreatment underwent dehydration to obtain HMF in first reactor which operated at 220 °C and 25 atm. HMF was introduced into the second reactor at 140 °C and 14 atm and then converted into levulinic acid, formic acid, and acetic acid, while the furfural and tar were generated from hemicellulose C5 sugars. Subsequent separation and purification steps were
carried out to gain yield pure levulinic acid. The byproducts (formic acid, furfural, tar formed after the reaction were separated away before recovering the main product. Finally, based on the simulated process, an EFB flowrate of 1000 kg/hour produces 146.9 kg/hour of levulinic acid.

2.1.3. Succinic acid production. Figure 1c shows a simple block diagram that was created to represent the process to produce succinic acid. This process route was based on experimental data from [16]. The reaction pathway to produce succinic acid involves the fermentation of C6 sugars at 39 °C for 29 hours. The reactor’s operating pressure were assumed to be at 1 atm since it was not specified in [16]. Dextrose, galactose and mannose were fermented to succinic acid via intermediate products, pyruvic acid and malic acid. Side reactions during the synthesis of succinic acid includes the production of carbon dioxide, lactic acid and acetic acid. Finally, 99.9 wt% of the succinic acid was recovered at the distillate at high purity of 99.9 wt%. To conclude, in the simulated process, an EFB feed flowrate of 1000 kg/hr produces 240.7 kg/hr of succinic acid. Thus, the overall process yield of succinic acid is 24 wt%.

2.2. Techno economic analysis
An economic viability of conversion EFB to both succinic acid and levulinic acid production can be assessed through techno-economic analysis (TEA). Equipment sizing and cost estimation cost employ Aspen process economic analyzer (APEA) V.10 and an investment factors to calculate project capital expenditures was taken from [17], as tabulated in Table 1.

| Component cost of investment | Factor |
|------------------------------|--------|
| **A. Total plant direct cost (TPDC)** |        |
| 1. Equipment purchase cost | 1      |
| 2. Equipment installation | 0.39   |
| 3. Instrumentation and control | 0.13   |
| 4. Piping | 0.31   |
| 5. Electrical | 0.10   |
| 6. Buildings | 0.29   |
| 7. Yard improvement | 0.10   |
| 8. Auxiliary facilities | 0.55   |
| **B. Total plant indirect cost (TPIC)** |        |
| 1. Engineering and supervision | 0.32   |
| 2. Construction | 0.34   |
| **C. Total plant cost (TPC = TPDC + TPIC)** |        |
| **D. Contractor’s fee and contingency (CFC)** |        |
| 1. Contractor’s fee | 0.05   |
| 2. Contingency | 0.10   |
| **E. Fixed capital investment (FCI = TPC + CFC)** |        |
| Working capital (WC) | 0.3    |
| Land use (LU) | 0.06   |
| **Total capital investment (TCI = FCI + WC + LU)** |        |

The construction time of project and working load of plant are set to be 3 years and 8000 hours per year. Working capital is set to be 30% of the total fixed capital investment due to the solid processing involved in biomass conversion processes. A 10% discounted cash flow rate of return is used over a 20-year plant life which has been often used for the small and medium plant sizes. The straight-line depreciation method is used for plant depreciation for 10 year. The annual production is formulated based on [18] which comprises of direct manufacturing cost (raw material cost, utilities cost, operating
labor, supervisory and clerical labor, maintenance and repair, operating supplies, laboratory charges, and patent and royalties), fixed manufacturing cost (local taxes and insurance and plant overhead), and general manufacturing cost (administration cost, distribution and selling cost, and research and development). The profitability analysis of both processes is performed by calculating the net present value (NPV), as stated in Equation (1).

\[
NPV = -TCI + \sum_{n=1}^{N_{\text{act}}} \frac{CF_n}{(1+i)^n}
\]

In this equation, CF denotes cash flow, i is the interest rate that assumed to be 10%, t is the plant life and TCI is total capital investment. The capacity of both processes is set to be 20 kton per hour of EFB. It can fulfill by 3% of global demand of levulinic acid in 2020 and by 50% of succinic acid in current production. According to current predictions, the levulinic acid market is expected to soar to 750 kilotons per year by 2020 [19]. Meanwhile, the current annual production capacity of bio-based succinic acid in the period of 2013 - 2014 is around 38,000 t that contributed 49% of the total market [20].

2.3. Life cycle assessment
Life cycle assessment methodology (LCA) is applied for quantifying the environmental impact. This approach has described in the ISO 14000 series of standards and has been employed extensively to assess the environmental impact for products and processes in bioenergy production systems [49]. An LCA is made up of four phases: (1) goal and scope definition, (2) inventory analysis, (3) impact assessment, and (4) interpretation.

2.3.1. Goal and scope. The aim of this study is to evaluate environmental impact of the process of converting EFB to levulinic acid and succinic acid. In order to be able to compare the both processes from empty fruit bunch, 1 kg output chemical obtained through each process is considered as functional unit. The selected system boundary is gate-to-gate which included acid hydrolysis, enzyme production, saccharification, levulinic acid, and succinic acid stage.

2.3.2. Life cycle inventory. The collection of inventory data is the crucial phase in performing LCA. The collection of input and output of mass and energy balance correlated to acid hydrolysis, enzyme production, saccharification, levulinic acid, and succinic acid stages which are then transformed into emissions and waste discharged. In this study, the emissions are considered as process direct emission, heat and electricity use in the system. Meantime, all emissions which come from electricity and heat generation are gained from Ecoinvent 3.4. The total life cycle inventory (LCI) can be expressed as a function of the emission process, the electricity and heat consumption, as given in Equation (2).

\[
LCI_{\text{tot}} = LCI_{\text{process}} + LCI_{\text{electricity}} + LCI_{\text{heat}}
\]

2.3.3. Life cycle impact assessment. The life cycle inventory is converted into the corresponding environmental impacts. In this case, the global warming potential (GWP) is taken as impact category measured in kg CO2-eq. The GWP is measured as the total of the GWP from each emission as seen in Equation (3).

\[
GWP = \sum_1 LCI_{\text{tot}} x m_i
\]

In this equation, \(GWP\) indicates the global warming potential while \(m_i\) is the damage factor that explains each greenhouse emissions are derived from [21].
2.3.4. Interpretation. The final phase of LCA where life cycle impact assessment (LCIA) result is elaborated and described to draw conclusions and decision-making in line with the goal and scope phase to acquire a more informed decision.

2.4. Inherent safety
The inherently safer approach or index-based approach is more suitable approach for hazard identification as it does not consume time, simple approach, easy interpretation, it provides net scores that can be used for hazard comparison to help decision-making, and it does not require the high level of expertise [22]. One of the index-based approach is hazard identification and ranking (HIRA) from [23]. It is developed to complement the shortcomings from previous approaches, Dow Fire & Explosion Index because it took into account more on expert opinion than on the system properties as well as it is a systematic and comprehensive approach that is easy to implement as well as it offers a reliable result [24].

The HIRA is essentially divided into two categories; the fire and explosion damage index (FEDI), and the toxicity damage index (TDI). The FEDI is calculated according to a thermodynamic model where there are five units that can be measured namely storage units, units involving physical operations, units involving chemical reactions, transportation units and other hazardous units. The TDI is a measurement of toxic load contained in the process unit.

The scope of this work is restricted to only main reactor of levulinic acid and succinic acid production. Penalty for the location of the nearest hazardous unit and space occupied by the unit are neglected because of lack of data. The available process data from simulation at early stage of process design are adequate for consequence analysis purpose. The information of HIRA calculation step and equation can be found in [23].

3. Results and discussion
3.1. Economic performance
The ability of the plant to operate at design capacity influences the economic viability of the facility. Figure 2 showed the comparison result of capital expenditure (Capex), operational expenditure (Opex), and yield from both levulinic acid and succinic acid. Overall the capital investment of levulinic acid and succinic acid were estimated at $35 million and $41 million, respectively. The total operating cost of levulinic acid and succinic acid were estimated at $19 million and $30 million, accordingly. The allocation of operating cost for each process can be seen in Figure 3, it was possible to see that a raw material and utilities costs presented a significant impact to the operating cost for both processes. In comparison, the succinic acid had higher raw material cost due to the expense of merchant CO₂ required by $1.3 million/year. Meanwhile, the utilities cost consisted of the electricity use and heat consumption which were responsible for the higher operating cost in succinic acid. It should be noted that the electricity use of succinic acid was higher by 42643 kWh, while levulinic acid was 1860 kWh. It caused the compressor unit for its CO₂ supply acid which contributed by 96% of total electricity use in succinic acid process. On top of that, the succinic acid consumed higher heat of consumption by around 211144 MJ/hour compared to levulinic acid with 135110 MJ/hour. The total revenue for levulinic acid and succinic acid were $195 million/year and $100 million/year, respectively. Then, NPV was calculated to evaluate the economic performance for both processes. The NPV for levulinic was found to be $1.1 billion in which nearly two-fold higher than succinic acid which was around $627 million. As a result, the levulinic acid showed a better result for economic performance compared to succinic acid. The financially attractive of levulinic acid was due to highly dependent on selling price of levulinic acid by 8.28 $/kg compared to succinic acid around 2.6 $/kg. Also, the operating cost was the factor boosting the economic performance of levulinic acid. It is interesting to note that levulinic acid posed the significantly lower of the process yield.
Figure 2. Capex and Opex of levulinic acid and succinic acid production.

Figure 3. Annual operating costs breakouts of levulinic acid and succinic acid production.

3.2 Life cycle assessment
The emissions associated to each activity for levulinic acid and succinic acid production were shown in Figure 4. These emissions were based on electricity use, heat consumption, and direct emissions of both processes. The emissions considered in this study included carbon dioxide (CO$_2$), carbon monoxide (CO), and methane (CH$_4$). Based on Figure 4, it is estimated that for every kilogram of levulinic acid produced, 6.3 kg CO$_2$-eq was released through levulinic acid process, whereas the total GWP of succinic acid was 11.2 kg CO$_2$-eq/kg succinic acid. Overall, the largest contribution of each process was coming from the stages of levulinic acid and succinic acid production themselves. Meanwhile, the acid hydrolysis, enzyme production, and saccharification only gave the paltry portion of contribution to the total GWP for both processes.

Overall, the electricity use in levulinic acid production contributed lower to the total GWP score than succinic acid production accounted for 4.4% and 33.2%, respectively. The more compressor used in succinic acid production resulted higher contribution to the total GWP score for electricity use. The heat consumption was primary contributor by more than 50% to total GWP for both processes. Another contribution of GWP was the direct emission for overall levulinic acid and succinic acid production. It could be observed from the Figure 5 that the direct emission from succinic acid production had the higher contribution to the total GWP than levulinic acid production. The unconverted CO$_2$ in succinic acid led to the higher contribution to the total GWP score. Thus, the levulinic acid production showed a better result for environmental impact evaluation from empty fruit bunch than succinic acid. Interesting to note that the heat consumption was being the major impact contribution to the GWP score for both processes.
Figure 4. Comparison of GWP of levulinic acid and succinic acid production.

Figure 5. The percentage contribution of GWP from electricity, heat, and direct emissions: (a) levulinic acid and (b) succinic acid.

3.3. Inherent safety measurement

The process aspects comprise of mass of chemical, operating conditions (temperature and pressure), and the chemical and physical properties information were considered as the essential data in the early process design phase. Data information used for analysis were reaction conditions, materials involved, heat of reaction, phase of reaction, units process involved, and process yield, transferred from Aspen Plus. In addition to, the information of chemical and physical properties was taken from safety data sheet (SDS). The two most hazardous chemical were identified for both processes namely furfural in levulinic acid production and succinic acid in its process. The furfural was a side product that formed from C5 sugars (xylose and arabinose). It categorized as toxic/corrosive based on NFPA ratings (NH = 4) and posed high flash point at 60 °C compared to other substances in levulinic acid process. Meanwhile, the succinic acid was selected as the most hazardous chemical. In comparison to others, it had got NFPA score (NH = 3).

According to the results shown in Table 2, as overall, the levulinic acid production showed higher results for both categories, FEDI and TDI, respectively. The Levulinic acid operated under severe operating conditions (200 °C, 14 atm), whereas succinic acid was in moderate operating conditions (39 °C, 1 atm). The physical energy, contained in levulinic acid, had a huge impact to the FEDI score. It was caused by higher temperature and pressure in reactor. The higher operating temperature and pressure posed a higher energy and risk for explosion and chemical leakage [22]. In comparison, the succinic acid revealed lower result of FEDI because of lower temperature and atmospheric pressure used in reactor.
Table 2. Comparison for FEDI and TDI score.

| Process unit   | Chemicals     | Type of hazard  | FEDI  | TDI  |
|---------------|---------------|-----------------|-------|------|
| Levulinic acid| Furfural      | Toxic/corrosive | 16.3  | 125.9|
| Succinic acid | Succinic acid | Toxic/corrosive | 7.4   | 3.08 |

The value of TDI of levulinic acid was higher than succinic acid even for FEDI values. The crucial points in TDI score were the pressure and vapor density. The vapor pressure was proportional to temperature, the high temperature would increase the vapor pressure by means the possibility of furfural changed into gas phase in case of leak would be higher. Hence, the succinic acid was inherently safer process design than levulinic acid.

4. Conclusions

Levulinic acid and succinic acid production were simulated using Aspen Plus and its sustainability performances of Net Present Values (NPV), Global Warming Potential (GWP), and Hazard Identification and Ranking (HIRA) were calculated accordingly. The comparison of both processes regarding the sustainability assessment was carried out. The results of flowsheet synthesis, process simulation, techno-economic analysis, and other sustainability considerations, such as life cycle assessment and inherent safety, were used to compare the upgrading pathways. It was found that levulinic acid showed better economic performance than succinic acid. Also, the levulinic acid showed a better result from environmental impact evaluation of 6.3 kgCO₂-eq/kg levulinic acid compared to succinic acid with 11.2 kgCO₂-eq/kg succinic acid. The safety assessment revealed that succinic acid was comparatively safer than levulinic acid. The severe operating condition played a major factor to FEDI and TDI score. In the future, the sustainability assessment of both processes can be integrated as biorefinery superstructure design where multi-objective optimization problem approach can be implemented.

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