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An integrated sustainable biorefinery concept towards achieving zero-waste production

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1. Introduction

Due to the ongoing climate crisis, the world is forced to search for renewable and green energy resources to meet the world’s growing energy demand (Mansouri et al., 2013, 2017; Vlysidis et al., 2011a). Al-Malah, 2019, the transportation sector in United States accounted for 28% of the total energy consumption (BP, 2020; EIA, 2020a). This sector is highly dependent on petroleum derivatives, which makes up 88% of its total energy consumption (EIA, 2020a; Ritchie and Roser, 2018). These petroleum derivatives pose an environmental challenge, as they are amongst main contributors to carbon dioxide (CO2) emissions contributor leading to the greenhouse effect and global warming (IEA, 2021, 2020). The threshold of 400 ppm-CO2 was passed in 2016 in atmosphere (Keeling et al., 2005); and 414 ppm has been passed by February 2020 (Lindsey, 2020). It is estimated that compared to pre-industrial levels, the human activities have caused an increase of 1 °C, in a likely range of 0.8 °C–1.2 °C (Masson-Delmotte et al., 2018). If it continues to increase within the current rate, the global increase is estimated to reach 1.5 °C between 2030 and 2052 above the pre-industrial levels (Masson-Delmotte et al., 2018). The environmental challenges for such increase could include severe weather and rising sea levels (Denchak, 2016). Production of bio-based fuels has become attractive in the past two decades (Sánchez-Ramírez et al., 2021; Segovia-Hernández et al., 2020).

Interest in biodiesel fuel (BDF) has been gaining attention (Mansouri et al., 2019; Vlysidis et al., 2011a) because it is non-toxic, renewable, sustainable, biodegradable, and has a very low concentration of sulfur (Zhang et al., 2003a). Furthermore, BDF emits ~70% fewer hydrocarbons, ~80% less CO2 and ~50% less particulates compared to petroleum fuels (Kiss and Bildea, 2012; Mansouri et al., 2013) Due to its environmental friendliness, multiple European Union directives and US incentives have been signed to replace diesel with BDF consumption (Kotrba, 2020). Its effect is evident, the increase in BDF consumption in US passed from 38 million-liters to 8 billion-liters between 2001 and...
2016, and in Europe from 5 billion-liters to 15 billion-liters between 2006 and 2016 (EIA, 2020b). Different type of fats can be used for BDF production (Hanson and Agarwal, 2018), such as primary vegetable oils, from soybean and canola, waste cooking oil (WCO) or animal fats (Hanson and Agarwal, 2018). The disadvantage of using vegetable oils is their high buying price and that they are first generation biomass, mainly intended for human consumption. Hence, making the problem both economical and ethical. In 2005, estimated soybean BDF production cost was 0.55 USD per diesel energy equivalent liter, whereas diesel wholesale prices averaged 0.46 USD/liter (Hill et al., 2006). In comparison to petroleum, BDF production can cost almost one and half times more, where the major cost is for raw materials (Lott, 2002; Prokop, 2002; Zhang et al., 2003a). Hence, as per the 2005 studies (Hanson and Agarwal, 2018), BDF is not cost-competitive with petroleum-based fuels, unless they are allocated subsidy. In US, subsidy of 0.29 USD/liter can be allocated, if the life-cycle environmental impacts of BDF are proved to be sufficiently less than petroleum fuel (Hill et al., 2006). Therefore, it is economically feasible to use WCO as feedstock, since it can be acquired at actively lower price of almost half of the oil prices (Krawczyk, 1999; NESTE, 2020; Zhang et al., 2003a). In addition to its lower cost, the process converts waste into a valuable product, as WCO is labelled as municipal waste (FUBIA, 2020) and has an estimated daily production of approximately 378 million-liters in the US alone, as indicated by Energy Information Administration (Panadare and Rathod, 2015). Such huge production of WCO would escalate the environmental risks if it is not properly disposed, including unpleasant odor, increasing level of organic pollutants in water and eutrophication (Azahar et al., 2016). If WCO is dumped in water without wastewater treatment, it is postulated that 1L of WCO can pollute up to 500,000L. (Panadare and Rathod, 2015). The use of WCO for BDF production would limit these environmental risks and offer a more sustainable choice and make the production eligible for subsidies. In 2017, only about 10% of the total BDF production was by WCO (Hanson and Agarwal, 2018), due to the complexity of gathering the WCO and the need of pre-treatments such as esterification and filtration for the high content of free fatty acids, moisture and presence of solid particles (Casallas et al., 2018; Thoai et al., 2019). As future perspective, BDF from third generation feedstock such as microalgae and oleaginous yeasts, currently under development, might be taken into consideration (Fidio et al., 2021).

Despite several environmental benefits, WCO cannot guarantee economically competitiveness, because of the processing and transport costs. One strategy for making BDF production more profitable is by transforming its waste streams into value-added products through process integrations based on the circular economy concepts aiming to eliminate waste and the continual use of resources (Mansouri et al., 2019; Monsivais-Alonso et al., 2020; Silk et al., 2020; Udugama et al., 2017; Vlysidis et al., 2011a). One such opportunity is in the production of glycerol (GLY), which is a by-product of BDF production. The increase in the production of BDF has negatively impacted the GLY market, and around 600 MMtons of crude GLY is produced annually (Gao et al., 2016). This oversupply has caused prices to plummet, making it a low-cost waste material (Fan et al., 2010). Instead of disregarding the crude GLY as a waste, it can be used as a feedstock for the production of bio-succinic acid (bio-SA) (Vlysidis et al., 2011b).

The bio-SA production is beneficial because it is capable of replacing 30 petrochemical products (Dickson et al., 2021; Mancini et al., 2019) and has a strong market value of 2.8 USD/kg (Pinazo et al., 2015; Tan et al., 2017). Further, bio-SA is produced through fermentation using CO2 and introduces carbon reduction (Li et al., 2018). Due to its lower toxicity compared to petroleum-based succinic acid, it is preferred in pharmaceuticals and cosmetics (Tan et al., 2017). Furthermore, its applications include 1,4-butadiol production, plasticizer, food additives, pigments and surfactants (Alexandre et al., 2019; Cavani et al., 2016; Mancini et al., 2019; Nghiem et al., 2017; Song and Lee, 2006; Vlysidis, 2011).

Another strategy for making the economics of BDF production more promising would be on-site methanol (MeOH) production due to the fact that 3 mol of MeOH are required to produce 1 mol of BDF. If MeOH produced from CO2 on-site becomes cheaper than its purchase, it could easily lower the production cost of BDF (Roh et al., 2016; Yang et al., 2018). The lower cost of MeOH production is an assumption and is tested through the economic analysis. Furthermore, the on-site MeOH production provides an opportunity to produce this chemical more environmentally friendly, because in industry, MeOH is synthesized from syngas, a mixture of hydrogen and carbon monoxide, obtained via two-step reforming of natural gas or gasification of coal (Dahl et al., 2014; Roh et al., 2016). This production method is associated with high emissions, whereas the process pathway used for this study uses CO2 as a raw material in combined reforming process (Frauzem, 2017; Roh et al., 2016). Hence, on-site MeOH production would ensure CO2 utilization, and this would allows reducing the carbon emission from the fossil-based power plants or cement plants, which would otherwise increase global warming (Roh et al., 2016). Therefore, integrating MeOH production with BDF production can make the overall process more economically and environmentally sustainable.

By the discussed points, an environmental friendly biorefinery (Fig. 1) can be designed and evaluated. This design and purpose aligns with the mission of the United Nations Framework Convention on Climate Change, which aims to minimize the carbon footprint through the renewable fuel production (UN, 2016).

In this study, it has been investigated how the BDF production can benefit from process integration with different chemicals. It is because as mentioned earlier, the biorefinery requires process integration to maximize their potential and for economic competitiveness with respect to petroleum refinery. Therefore, four scenarios for an integrated biorefinery are investigated and evaluated in terms of profitability and sustainability to investigate whether the integration through circular economy offers any benefit. More specifically, the study includes the analysis of one stand-alone and three integrated scenarios: (1) stand-alone BDF production; BDF production integrated with (2) MeOH production; (3) bio-SA production; and (4) MeOH and bio-SA production. The process pathway for each process was taken from literature (Dickson et al., 2021; Freres, 2016; Li et al., 2018; Mansouri et al., 2013; Roh et al., 2016) and the rigorous mass and energy balances were obtained by Aspen Plus V11 simulations. Each scenario was then evaluated based on rigorous profitability, sensitivity and environmental analysis using ECON (PSE for Speed, 2020), SustainPro (Carvalho et al., 2013) and LGSoft (Rattanatum et al., 2018). Upon identifying the bottlenecks and limitations from analysis, new sustainability goals were set and implemented to investigate the improvement in economics and environmental sustainability. To this end, this approach assesses whether the process integrations based on circular economy concepts can allow moving towards a zero-waste and carbon-neutral biorefinery while staying profitable compared to a stand-alone BDF production.

2. Process descriptions: biodiesel, bio-succinic acid, and methanol from CO2

The process description and the procedure for conducting the Techno-economic analysis (TEA) and Sustainability analysis are discussed below.

2.1. Process description

The processing pathways for the production of BDF (Mansouri et al., 2013), MeOH (Frauzem, 2017; Roh et al., 2016), and bio-SA (Dickson et al., 2021; Freres, 2016; Kurzrock and Weuster-Botz, 2010; Vlysidis et al., 2011a) were gathered from superstructure optimizations from literature because it provides the benefit that all the processes have already been optimized and optimal processing routes are identified. For BDF production, an enzymatic process pathway is used, which utilizes WCO and MeOH. The benefit of using enzymatic pathway is that it
allows the reaction conditions to be kept at ambient conditions, hence minimizing the risk and operating costs. The combined reforming process is used for MeOH production (Frauzem, 2017; Roh et al., 2016), while engineered E. coli is used as microbial host for bio-SA production (Freres, 2016; Kurzrock and Weuster-Botz, 2010; Li et al., 2018; Vlysidis et al., 2011a). All three processes were simulated in Aspen Plus V11 (Freres, 2016; Kurzrock and Weuster-Botz, 2010; Li et al., 2018; Vlysidis et al., 2020; Rasool Lone and Ahmad Rather, 2015).

2.1.1. Biodiesel production

The inlet streams consist of WCO stream consisting of triglycerides and free fatty acids, and MeOH with a purity of 99 w/w% (Mansouri et al., 2013). As shown in Fig. 2, the inlet stream of WCO is heated (E-100) to 95 °C/1 bar and mixed (MIX-103) with MeOH stream and sent to the transesterification reactor (R-101) with temperature of 69 °C/1 bar. Inside the reactor, a three-stage transesterification of triglyceride, diglyceride, and monoglyceride with MeOH of 0.3 kg/kg-acids takes place (Mansouri et al., 2013). The MeOH is used in excess to move the reaction towards the products side. The second part of the reaction involves esterification, which is applied to reduce the free fatty acids in WCO (Mansouri et al., 2013). The reactor produces BDF as a main product and GLY as a by-product, with unreacted MeOH and water. The reactor outlet stream is pumped (P-103) and heated (E-101) to 162 °C and sent into a flash tank (F-101) that separates MeOH/water mixture from BDF/GLY mixture. The MeOH/water mixture is cooled down to 62 °C in a cooler (E-102) and passed through a 3-phase separator (V-100) before being sent to a 22-stages distillation column (T-101). The column purifies MeOH to 99.99 w/w%, making it eligible for recycling. The MeOH is cooled down (E-103) and recycled. Meanwhile, the BDF/GLY stream is cooled down to 12 °C by a cooler (E-105) and sent into a 3-phase separator (V-101), which produces crude BDF and crude GLY. The crude GLY stream is brought to 220 °C using cooler (E-105) and it is sent into a flash tank (F-102), which produces 99.8 w/w% GLY stream. The vapor containing MeOH is cooled down in a cooler (E-106) and sent to the 12-stages distillation column (T-102). The 99.96 w/w% purified MeOH is cooled (E-107) and recycled. The crude BDF stream from the 3-phase separator (V-101) is sent into a 5-stages distillation column (T-103), which separates the BDF from MeOH, and the MeOH is recycled. The bottom stream containing BDF and waste oils is sent into a 41-stages distillation column (T-104), which purifies the BDF (methyl-esters), while waste oils (mono-, di-, and tri-glycerides) are discarded (Mansouri et al., 2013).

2.1.2. Bio-succinic acid production

The Bio-SA crystals are produced using GLY as a carbon source with water, ammonia, and CO₂ added as raw materials (Carlson et al., 2016; Li et al., 2018; Nejadfomeshi, 2013). The inlet streams at 25 °C/1 bar are mixed (MIX-302) and heated (E-301) to 37 °C before fermentation (R-301). The microbial host for the fermentation is an engineered E. coli (Li et al., 2018), which produces bio-SA and acetic acid, and biomass (DCW) and water as by-product (Li et al., 2018). The operating conditions for the fermentation are 37 °C/1 bar and residence time is 72 h (Li et al., 2018). The fermentation is pH sensitive, therefore sodium hydroxide is added as buffering agent, and produces sodium succinate (Morales et al., 2016). The fermenter outlet stream is passed through a flash tank (F-301) to remove the gases (Li et al., 2018). The first separation step is microfiltration followed by nanofiltration, for which the operating temperature and transmembrane pressure are 45 °C/1 bar and 30 °C/11 bar, respectively (Freres, 2016; Kurzrock and Weuster-Botz, 2010; Thuy and Boontawan, 2017). The operating conditions for the filtrations are achieved by pumps and heat exchangers, P304/E303 respectively. The next step is the acidification (R-302) at 25 °C/1 bar using sulfuric acid to recover free bio-SA and reduce pH for crystallization (Cheng et al., 2012; Lubsungneon et al., 2014). The outlet stream is cooled (E-305) and sodium sulfate is filtered (FIL-301) and dried (F-302) to 98 w/w%. The free bio-SA stream is concentrated to 19% bio-SA solution by passing it through an evaporator (F-303) operating at 100.9 °C/1 bar (Morales et al., 2016). The evaporated water is 99w/w% pure and vapours are cooled down (E-307) to 25 °C, purged (SP-301) and recycled to the fermenter. The concentrated bio-SA stream is cooled to 4 °C (E-308) and sent to a crystallizer (CR-301) (Morales et al., 2016). The bio-SA is crystallized and the crystals are filtered (FIL-302) and dried (F-304) and purity of 99.6% are recovered (Freres, 2016; Morales et al., 2016). During the purification and recovery, 16% of bio-SA is lost.

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Fig. 1. Principles of Circular Economy for designing a zero-waste biorefinery.
2.1.3. Methanol production from CO₂

The feedstock used for the methanol production is methane (CH₄), CO₂, and steam (Frauzem, 2017; Roh et al., 2016). The incoming gases are compressed to 25 bars by C-201 and C-202 for CH₄ and CO₂ respectively. The incoming water stream is heated (E₂01) to produce steam and compressed (C-203) The three gases are mixed (M-200) and heated to 915°C (E₂02 and E₂03) before the combined reforming reactor (R-201). The combined reforming is modelled using a Gibbs Reactor, operating at 915°C/25 bars, which reduces the Gibbs free energy and produces syngas with a 2H₂:1CO ratio (Roh et al., 2016). The syngas is mixed with the recycling stream (M-201), compressed (C-203) and heated (E-204) before sending it to reactor (R-202) operating at 240°C/60 bars, where MeOH is produced (Frauzem, 2017; Roh et al., 2016; Vanden Bussche and Froment, 1996). After the synthesis, the stream is cooled (E-205) and the light components are removed using a flash tank (F-201) operating at 60°C/60 bars, and 35% of the vapor stream is purged (SP-201) while the rest is recycled (Frauzem, 2017). After flashing, additional light components are removed using a second flash tank (F-202) operating at 145°C/11 bars, so that inert components do not blanket the condenser (Frauzem, 2017; Luyben, 2010) The stream is then sent into a distillation column (T-201) to separate the MeOH-WAT mixture. The column produces a 99.6 w/w% pure stream of MeOH (Roh et al., 2016).

2.2. Process integration

In process integration, MeOH produced is used as a feedstock for the BDF production and the GLY produced from BDF is used for the production of bio-SA. The process flow diagram for each scenario is shown in Fig. 2, where the dotted line represents the process integration. The mass and the energy balance is shown in Fig. 3.

2.3. Techno-economic analysis

Techno-economic analysis (TEA) is performed to assess the profitability of all four scenarios, using the cost model from Luyben (2010) and Peters and Timmerhaus (1994). The assumptions considered are: (1) the operating hours are 8400 h/yr (350 days) (Mansouri et al., 2013);
(2) plant lifetime is 20 years; (3) all prices are in USD for year 2019; (5) it takes 3 years to build the plant where 15%, 35%, and 50% of Fixed Capital Investment (FCI) is used in the first three years respectively; (6) plant reaches its full capacity in the second year (Peters and Timmerhaus, 1994); (7) there are no extra investment during the 20 years (Zhang et al., 2003a); (8) the inflation rate is 0 for both total production cost and annual sales; (9) 30% income tax (Peters and Timmerhaus, 1994); (10) 10% net rate of return (Peters and Timmerhaus, 1994); (11) depreciation is accounted using the MACR method (Peters and Timmerhaus, 1994); and (12) land cost is taken as 6% of the total installed cost (Davis et al., 2015).

2.3.1. Capital investment and annual cost

The total capital investment (TCI) is the total investment required to erect a full functioning plant (Peters and Timmerhaus, 1994). It was calculated as the sum of fixed capital investment (FCI), working capital investment (WCI), and land cost (LC) (Zhang et al., 2003b). They are all calculated from the total installed cost (TIC), which is the total cost of all equipment after installation and is the sum of each equipment’s installed cost (IC) (Equation (2)) (Vlysidis et al., 2011a). The IC for each equipment is calculated by (1) scaling index to scale the equipment (Toby, 2018), (2) installation factor (IF) to compute installation cost (Toby, 2018), and (3) chemical plant cost index (CEPCI) to update the cost of equipment from the base year to the analysis year (2019) (Equation (1)) (Vlysidis et al., 2011a). The 2019 CEPCI value was 607.5 (Chemical Engineering, 2020).

\[
IC = \left( \frac{\text{New Capacity}}{\text{Base Capacity}} \right)^n \cdot \text{Base Cost} \cdot \text{CEPCI} \cdot \text{IF} \tag{1}
\]

\[
\text{Scaling Law}
\]

\[
TIC = \sum_{i=0}^{n} IC_i, \text{ where } a \geq 0 \tag{2}
\]

The base cost is the literature cost for base capacity from base year, and \( n \) is the scaling exponent (Davis et al., 2015; “Equipment Cost,” 2020; Tan et al., 2015; Toby, 2018). Detailed explanation for the equation is provided in supplementary material. As the processes were simulated within Aspen, therefore the TIC for the flash tanks, distillation column and heat exchanger was taken directly from the Aspen’s Economic Analyzer. The total manufacturing cost (TMC) is the annual production cost and takes into account the raw material, utility, waste disposal, carbon tax, and extra costs essential for the operation of the plant (Peters and Timmerhaus, 1994; Tan et al., 2015). The extra costs include maintenance repairs, operating supplies, etc. (Peters and Timmerhaus, 1994). The utility cost includes the cost of steam, electricity, refrigerant, and cooling water. High (HP) and low-pressure steam (LP) is used for heating and cooling water and refrigerant is used for cooling. The waste disposal cost is the penalty price depending on whether a waste is toxic or non-toxic, and penalty were found to be 213.2 USD/ton and 32.0 USD/ton, respectively (Peters and Timmerhaus, 1994; Vlysidis et al., 2011a). The carbon tax is a penalty which has to be paid for burning of fossil-based fuels such as coal, oil and/or gas (CTC, 2020). In this study, fossil fuel is only used as a fuel source to produce the utilities, therefore, the carbon tax is not applied for the direct CO\(_2\) emissions, but only for the indirect CO\(_2\) emissions and is 15.0 USD/ton (Bolle, 2019; CTC, 2020). The cost of utility for electricity, HP steam, LP steam, refrigerant and cooling water are 16.1 USD/GJ, 5.8 USD/GJ, 2.6 USD/GJ, 4.4 USD/GJ and 0.4 USD/GJ respectively (EIA, 2020c; Al-Malah, 2019; Intratec, 2020; Sinnott and Towler, 2009; Turton et al., 2009).

Fig. 3. - Mass and energy balance for the integrated biorefinery.
2.3.2. Profitability criteria

Economic metrics including net present value (NPV), return on investment (ROI), discounted cash flow rate (DCFR), and payback period (PP) were calculated in order to evaluate the profitability of the scenarios (Chen and Mansa, 2020a, 2020b; Kagan and Drury, 2020; Kenton and Mansa, 2020).

2.3.3. Sensitivity analysis

The single point sensitivity analysis was performed for each scenario to investigate the impact of selected parameters on profitability (NPV). The selected variables, shown in Table 5, are the ones expected to change within the plant lifetime. The raw material and product price are the most probable to deviations because of the changing international market. The economic assessment is performed for a theoretical plant in 2019, but if the plant is to be built in the future, then the equipment cost may change, which directly changes the TCI (Bolle, 2019). The utility cost may also change due to the change in fuel prices. For the perturbations for each of these parameters, a tornado plot for NPV was plotted and analyzed (El-Temtamy and Gendy, 2014; Lee and Rim, 2016).

2.4. Sustainability analysis

The sustainability analysis was performed using SustainPro (Carvalho et al., 2013) and LCSoft (Rattanatam et al., 2018). In order to recognize the process bottlenecks and carbon footprint, respectively. SustainPro uses the mass and energy balances and the methodology from Carvalho et al. (2013) to identify the open paths (OP) and cycle paths (CP). The OP are the paths through which the products and energy leave the system boundary, whereas the CP are the paths through which the material and energy remain within the boundary in form of a loop (Carvalho et al., 2013). For the identified path, indicators such as total value added (TVa), material value added (MVA), and energy waste cost (EWC) were computed (Carvalho et al., 2013). The MVA indicates the value added between the entrance and exit of a given compound and is thus only calculated for OPs, while EWC reflects the costs due to utility consumption and waste treatment of a component path flow for OP and CP (Ng et al., 2015). The TVa relates the two indicators and shows the economic impact of a given path for the compound (Ng et al., 2015). A negative MVA means that the value is being lost as the compound enters and leaves the process, whereas a negative EWC means a high energy consumption and a negative TVa indicates that both the compound’s value and energy along the path is being wasted (Carvalho et al., 2013; Ng et al., 2015). Based on these indicators, the OP and CP which act as the bottlenecks were recognized, for further process intensification and improvement (Mansouri et al., 2013). LCSoft uses the utility requirements to compute the carbon footprint and indirect CO2 emission (Rattanatam et al., 2018). Since Scenario 2 and 4 have CH4 emission, which is a greenhouse gas therefore the CO2-equivalent (CO2-Eq) was calculated using the global warming potential (GWP) factor of 84 kg-CO2/kg-CO2 for a period of 20 years (IPCC, 2015). This is done to evaluate the global warming impact of greenhouse gases, and the GWP conversion is the measure of warming that is contributed by the greenhouse gas to the greenhouse effect equivalent to 1 kg of CO2 (CBS, 2019; IPCC, 2015). Hence, this conversion computes the amount of CO2 being released in terms of CH4 emission. Lastly, the carbon reduction for each process was found as the percentage difference between the CO2 input and output.

2.5. Base case design and improved design

The analysis are divided into two parts: base case design and improved design. In the former, the TEA followed by sensitivity analysis and environmental sustainability analysis were performed to assess the economic viability, bottlenecks, and environmental impact of all scenarios. Based on the analysis, the best-identified base case scenario was selected and new targets were set, and the process was simulated and analyzed from a profitability and sustainability viewpoint to investigate improvements.

3. Results and discussion

In this study, using a circular economy approach, four biodiesel based biorefinery scenarios (Table 1) were designed and analyzed to assess whether it can lead to better economics and a zero-waste scenario. Both the base case design and the improved design are discussed below in terms of economics and sustainability.

3.1. Base case design

Comprehensive simulations were performed to assess the technological feasibility of process scenarios and to obtain rigorous material and energy balances for profitability and sustainability analysis. Results in Table 2 show that all scenarios require 366 MM/kg/yr of WCO to produce 358 MM/kg/yr of BDF. Scenario 1 has the least raw material requirement, whereas Scenario 2 to 4 have high raw material requirements, in increasing order, due to process integrations. In terms of value-added products, Scenarios 1 and 2 produce biodiesel as the main product and GLY as a by-product. The advantage of Scenario 3 and 4 is production of a high market value product i.e, bio-SA with two times higher market value than that of BDF. Hence, by doing so, the revenues are increased.

In terms of energy requirement, Scenario 1 consumes 2.6 GJ/kg-products, where 62% of total energy is consumed in heating and remaining in cooling. In Scenario 2, the energy requirement increased to 5.0 GJ/kg-product – almost double than that required in Scenario 1, due to the elevated temperature (915 °C) and high compression (60 bar) in
the reactor in MeOH production. Scenario 3 has an energy requirement lower than that of Scenario 2 but almost double as much for Scenario 1. The biggest energy contributor in Scenario 3 is the heating. Scenario 4 is complete integration of all three scenarios, and has the highest energy requirement, which is 220%, 48% and 57% more than the Scenario 1, 2, and 3, respectively.

Overall, results indicate that the process integration increases the material and energy requirements compared to the standalone process. In contrast, process integrations produce multiple end-products of high value that may increase the overall profitability and sustainability. Therefore, in the subsequent sections, rigorous profitability and sustainability analyses are performed to determine if circular economy concepts are achieved effectively.

3.1.1. Techno-economic analysis

TEA results in Table 3 indicate that Scenario 1 has the least capital cost of 42.8 MMUSD, whereas the capital cost increases from Scenario 2 to 4 in an increasing order, i.e., 2.3, 3.2, 4.6 times higher than Scenario 1. The integrated scenarios are capital intensive due to addition of processing areas needed to utilize waste streams generated from standalone BDF process. From Total Capital Investment (TCI) breakdown of each scenario (Fig. 4A), the purification area in Scenario 1 is the main consumer of TCI, which is due to purification of the crude BDF by using 5 distillation-columns. In Scenario 2, integration of MeOH production with BDF process, consumes 56.7 MMUSD of additional investment, in which maximum investment is used in pre-treatment area. Interestingly, storage cost in Scenario 2 is found to be higher than others, which was due to large storage of CO2 for the MeOH process. The Scenario 3 has the second highest TCI, due to the fermentation costs. The fermentation in bio-SA has a residence time of 72 h and a high requirement of water due to low titer (g/L), making the size requirement high (Li et al., 2018). In order to accommodate that, a total of 9 fermenters of 500 m³ capacity are being used, bringing the fermenter cost at 15 MMUSD. Whereas in BDF and MeOH, the residence time for their reactions are relatively lower, therefore the reactor volumes are smaller and cheaper (Mansouri et al., 2013; Roh et al., 2016). Scenario 4 has the highest TCI that requires additional investment of 152.7 MMUSD compared to Scenario 1.

As shown in Table 3, the highest Total Manufacturing Cost (TMC) is from Scenario 4, followed by Scenarios 2, 3, and 1. As further shown by Fig. 4B, in all scenarios, the main cost driver for the TMC is the raw material cost. The main contributor for the raw material in each scenario is the WCO, which has a unit price of 0.2 USD/kg (Farid et al., 2020). Since it has a high mass requirement of 366 MMkg/yr, the final expenditure becomes 73.2 MMUSD/yr in each scenario. Scenario 2 has a raw material cost of 106 MMUSD/yr, in which 33 MMUSD/yr is used for the MeOH production. In MeOH production, the main contributor to raw materials cost is the methane purchase, costing 28 MMUSD/yr, which is a major bottleneck. This is because the total external purchase cost of MeOH in Scenario 1 is 13 MMUSD/yr, while in Scenario 2 only methane costs 28 MMUSD/yr, which is 15 MMUSD/yr higher than purchasing only MeOH. Already here, the expenditure of MeOH production exceeds that of Scenario 1. The second weakness in Scenario 2 is that all the raw materials have to be bought externally and methane is the most expensive raw material among all. While in Scenario 3, there is a cost saving involving GLY, as it is the feedstock with the highest mass requirement but is being produced in-house and does not have to be bought. At the same time, the other raw materials required for bio-SA production are relatively cheaper, giving a total expense of 4.2 MMUSD/yr. Therefore, the TMC for Scenario 3 is lower than Scenario 2. The Scenario 4 has the highest amount of raw materials which need to be bought, thus that expenditure is the highest.

The second dominating parameter in the TMC is the utility expenditure, which includes both waste and utility costs. The utility distribution is not very dominant in Scenario 1, but has a major role in Scenario 2, 3, and 4 as shown by Fig. 4B. The benefit of Scenario 1 is that the stand-alone BDF production is an enzymatic process and the reactions take place at room temperature, consequently, requires lower utility. Additionally, there is a relatively lower waste disposal production compared to other processes. Hence for Scenario 1, in the total utility cost of 7 MMUSD/yr, the HP steam, LP steam, cooling water, hazardous and non-hazardous waste disposal makes up 65%, 7%, 3%, 16% and 9% respectively. There is a hike of 265% in utility cost for Scenario 2 compared to Scenario 1. Scenario 2 accounts for 26 MMUSD/yr utility cost, while the MeOH production is responsible for 19.2 MMUSD/yr. In its utility cost, HP Steam, Electricity, cooling and hazardous waste disposal, makes up 22%, 19%, 1% and 55% respectively. Unlike BDF production, the high cost in MeOH is due to the high requirement of HP steam and electricity because all the reactions and compressions occur at high temperature and pressures. Furthermore, MeOH production has a high emission of hazardous chemicals such as carbon monoxide (4497 kg/h) and methanol (484 kg/h), while BDF had a total production of 635 kg/h hazardous waste. Non-hazardous waste was more prevalent in BDF production (2467 kg/h), which was still lower than the hazardous waste production in MeOH. Since the cost of hazardous waste disposal is almost 85% higher than non-hazardous waste, it makes MeOH production more expensive. In Scenario 3, the utility expenditure is 46% less than Scenario 2 but double that of Scenario 1. In Scenario 3, in contrast to MeOH process, the bio-SA process only adds an additional cost of 6.7 MMUSD/yr on top of the BDF production. For bio-SA, the highest cost comes from use of LP Steam and wastewater disposal, making up 31% and 32% of the total utility cost. In addition to these, the cooling, refrigerant, non-hazardous and hazardous waste disposal makes up 4%, 3%, 8% and 22%. Similar to BDF production, the bio-SA production takes place at low temperature and pressure (<37°C/1 bar), thus we need to buy steam or electricity. The only use of steam is for the evaporator. There is much higher need for cooling water in the bio-SA process, but since this energy utility is the cheapest, therefore the total utility cost does not increase much. In contrast to MeOH process and similar to BDF process, bio-SA production is more prevalent in non-hazardous waste production (2050 kg/h) but has a slightly higher hazardous waste production (810 kg/h) than BDF. The main disadvantage of the bio-SA process is its water disposal costs of 2 MMUSD/yr at a unit price of 0.0132 USD/kg, the highest among all the
processes (Saltworks, 2019). It is because 18 kg-water/kg-GLY is required to dilute the GLY before fermentation due to low titer (g/L), and after fermentation this water become dirty, which takes high separation and purification costs. Hence, the main reason for the high utility cost in Scenario 2 is high power, HP steam, and the hazardous waste disposal, whereas in Scenarios 1 and 3, the entire production takes place at low temperature and pressure and has high non-hazardous waste disposal, keeping the utility cost lower. Whereas the Scenario 3 is only detrimental due to its high wastewater disposal cost compared to Scenario 1 and 2. Scenario 4 has the highest utility requirement due to the fact that it entails weakness of all other scenarios.

The total annual sales are the same for Scenarios 1 and 2 and for Scenarios 3 and 4 (Table 3) (Fig. 4C). In the first two scenarios, the sales come only from selling BDF and GLY, but the main revenue comes from BDF sales, which stands at 331 MMUSD/yr. Whereas in Scenario 3 and 4, sales are increased due to addition of bio-SA and sodium sulfate, as value-added by-products. Due to these two products, the sales increase by 78 MMUSD/yr, and hence removing the burden from BDF price.

Profitability results (Table 4) for Scenario 2 show that the NPV decreased by 25% compared to Scenario 1, due to higher capital and TMC. Despite additional expenses, Scenario 2 does not produce additional value-added by-product, which kept the income constant to that achieved in Scenario 1. In addition, on-site methanol production is not found economically favorable in respect to its purchase. Consequently, adding further expenses in Scenario 2 onto the constant income causes the NPV to decrease and the minimum selling price for BDF to increase. In contrast, the NPV in Scenario 3 increased because Scenario 3 produces two additional value-added by-products and gains higher revenues from selling multiple products in comparison to Scenario 2. Although capital and TMC significantly increase in Scenario 3, these additional sales cover the expenses of the new integrated process in most profitable manner. Therefore, the minimum biodiesel selling price also decreases. In Scenario 4, the new expenses are more dominant than the sales, thereby, the NPV decreases and the minimum BDF selling price increases. By comparing the NPV and minimum biodiesel selling price of all scenarios, it can be concluded that Scenario 2 is economically the worst scenario while Scenario 3 is the most optimal scenario.

![Fig. 4. Relative distribution of total capital investment (A), annual manufacturing costs (B), and annual sales (C) for each scenario in base case design.](image-url)
3.1.3. Sustainability analysis

The single-point sensitivity analysis was performed for each scenario to investigate the impact of biorefinery parameters on profitability (NPV). The parameters for which the sensitivity analysis was performed are provided in Table 5 along with their variations, and the resulting tornado plots are shown as Fig. 5.

For all the scenarios, the BDF price had the strongest effect on the NPV, because the entire profitability is dependent on its price. The second sensitive variable is the WCO price, because as discussed earlier, it is biggest expense and is common in all scenarios. Therefore, if its price changes, the NPV is greatly affected. For all scenarios, the income tax showed a huge impact on the NPV, which reiterates the idea that the tax exemption on the green production does lead to a higher NPV and profitability (Hanson and Agarwal, 2018; Hill et al., 2006). Scenario 1 and 2 have no bio-SA sales, therefore the price deviation in bio-SA has no influence in these two, but in Scenario 3 and 4, the bio-SA has a great influence on the NPV. This is because the sales from bio-SA is relatively high and plays a major role in streamlining the profitability, therefore if the bio-SA changes, it directly puts pressure on the BDF price. The utility and TCI did not influence the NPV much in Scenario 1, possibly due to their low values. But in the case of Scenario 4 and 3, these variables were much more influential. For the methane prices, it is the least influential in all of the scenarios. In conclusion, it can be stated that the BDF price, WCO price, and income tax are the top three variables which have the biggest influence on the NPV. Meanwhile, methane price and total utility cost has the lowest influence on the NPV.

3.1.4. Sustainability analysis

For the sustainability analysis, using SustainPro, the Open (OP) and Closed paths (CP) were identified and evaluated (Carvalho et al., 2013). Results in Table 6 show that among all the compounds, water in Bio-SA process has the lowest Total Value Added (TVA) followed by MeOH in BDF process. The negative TVA values mean that the paths have the highest material loss and energy consumption. Hence, all of these open paths are the bottlenecks and must be improved in order to recover the valuable materials in both raw material and energy. Furthermore, Table 6 shows that MeOH, CO₂ and water have the highest Energy Waste Cost (EWC) values in a closed-path, which are the recycle paths. These paths indicate which unit operations are the potential bottlenecks (Mansouri et al., 2013).

As shown in Fig. 6, in terms of energy per product, the highest requirement is for Scenario 4 followed by Scenario 2. The product is defined as the total mass of chemical being sold ahead for profit. The reason for the high energy requirement is the MeOH process, as discussed earlier. In Scenario 1, the total waste production is 0.07 kg/kg-product, and it includes 0.05 kg/kg of non-hazardous waste such as waste oils. In Scenario 2, the waste production increases to 0.36 kg/kg-product, of which 0.14 kg/kg and 0.08 kg/kg correspond to hazardous waste and wastewater. As it was discussed earlier, for this spike is the production of CO in the MeOH process. In Scenario 3, the waste production is at 0.68 kg/kg-product, and it includes 0.05 kg/kg of wastewater and non-hazardous waste. The scenario 4 has the highest waste production owing to all of the aforementioned waste types. Thus, the strength of Scenario 1 and 3 is that they produce non-hazardous waste, whereas the biggest weakness of Scenario 2 and 4 is that there is a high production of hazardous waste. Even though the waste production of Scenario 3 is higher than Scenario 2, but the environmental impact of Scenario 2 is more detrimental than Scenario 3. As the CO emission leads to greenhouse effect, while the wastewater can be purified and recycled back into the process. Therefore, in terms of waste production, the Scenario 1 and 3 are preferred.

Two emissions from biorefinery should be considered: direct and indirect emissions. The direct emissions result from releasing the pollutants into the environment, while the indirect emissions result from burning fossil fuels to generate utility for biorefinery. In terms of indirect CO₂ emissions, Scenario 1 has the lowest (0.02 kg-CO₂/hr) emissions due to low fossil fuel being used to generate utility for the process. Meanwhile in Scenario 2, the indirect CO₂ emissions increase to 1.06 kg-CO₂/hr due to the high utility employment in the MeOH process. The highest emissions in the MeOH are from the electricity, followed by heating and then cooling. In Scenario 3, the emission was 0.04 kg-CO₂/hr, which is doubled respect to Scenario 1, but almost three times less than Scenario 2. The final CO₂ emissions in Scenario 4 are 1.09 kg-CO₂/hr.

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**Table 5**

<table>
<thead>
<tr>
<th>Variables</th>
<th>Unit</th>
<th>Min</th>
<th>Base</th>
<th>Max</th>
<th>Variation (%)</th>
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</thead>
<tbody>
<tr>
<td>Biodiesel price ($/kg)</td>
<td>0.69</td>
<td>0.92</td>
<td>1.15</td>
<td>±25</td>
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</tr>
<tr>
<td>Waste Cooking Oil price ($/kg)</td>
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<td>0.20</td>
<td>0.25</td>
<td>±25</td>
<td></td>
</tr>
<tr>
<td>Succinic Acid price ($/kg)</td>
<td>2.49</td>
<td>2.93</td>
<td>3.37</td>
<td>±15</td>
<td></td>
</tr>
<tr>
<td>Methane price ($/kg)</td>
<td>0.66</td>
<td>0.77</td>
<td>0.89</td>
<td>±15</td>
<td></td>
</tr>
<tr>
<td>Income Tax rate (%)</td>
<td>27</td>
<td>30</td>
<td>33</td>
<td>±10</td>
<td></td>
</tr>
<tr>
<td>Total Utility cost</td>
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<td>DFES</td>
<td>DFES</td>
<td>DFES</td>
<td>±20</td>
</tr>
<tr>
<td>Total Capital Investment</td>
<td>MMS</td>
<td>DFES</td>
<td>DFES</td>
<td>DFES</td>
<td>±20</td>
</tr>
</tbody>
</table>

DFES: Different for each Scenario.

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**Table 6**

Selected Variables for Sensitivity Analysis and their respective perturbations (Rolle, 2019; Peters and Timmerhaus, 1994).

<table>
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<tr>
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<td>DFES</td>
<td>DFES</td>
<td>±20</td>
</tr>
</tbody>
</table>

DFES: Different for each Scenario.
Hence, in conclusion on the basis of the sustainability metrics, it can be stated that the Scenario 3 is the most optimum owing to its lower waste production, energy requirements and the indirect CO$_2$ emissions.

3.1.4. Life cycle assessment

In terms of the life cycle assessment, Table 7 indicates that Scenario 1 offers no carbon reduction. Scenario 2 has a carbon reduction of 67% (without CO$_2$-eq of CH$_4$) and 79% (with CO$_2$-eq of CH$_4$). It has the highest carbon and CH$_4$ input from the MeOH process, and hence main output is the CO$_2$-eq of CH$_4$ followed by the direct emission. In terms of the indirect emission, MeOH process has 5 unit operations which have a footprint of more than 0.5 kg-CO$_2$/hr. Scenario 3 has 90% carbon reduction, but compared to Scenario 2, it has 86% less CO$_2$ input, because CO$_2$ is used only for the microorganism growth rather than as a stand-alone raw material, additionally there is no CH$_4$ input or emission, hence no CO$_2$-eq. Scenario 3 also has 96% less direct and indirect emission of CO$_2$ compared to Scenario 4.

3.2. Improved design

Upon performing the TEA and sustainability on the base case design, new targets were set in order to achieve a more favorable scenario in terms of economics and sustainability. From the initial analysis, it was concluded that Scenario 3 was the most optimal scenario in terms of both economics and sustainability.

Many earlier studies have investigated the economic potential of BDF production (Apostolakou et al., 2009; H. West et al., 2008; Marchetti and Errazu, 2008; Mizik and Gyarmati, 2021; Singhabhandhu and Tezuka, 2010; Zhang et al., 2003b, 2003a). The studies have highlighted that BDF is not cost competitive to petroleum fuel, due to the high cost of raw materials and production. The only possible method to increase the profitability is through converting by-products into profitable products (Vlysidis et al., 2011b). The Scenario 3 investigated in this study is more environmental friendly and eligible for tax exemption (Hanson and Agarwal, 2018).

Therefore, given the benefit and relevance of Scenario 3 over Scenario 1, modifications were made to the Scenario 3 and for the new design, the simulations were run and TEA and sustainability was performed.
3.2.1. Process design

For the improvements, modifications were made to the bio-SA production process. In terms of indirect CO₂ emission, the highest indirect emission in the process was due to the high energy demand by the evaporator. Therefore a multi-effect evaporator (MEE) was added to the process. It was set up in a way that the inlet stream before evaporation was divided into three streams. The first stream is heated up to 101.92 °C using steam. The vapours from the first evaporator are sent into a heat exchanger in order to heat up the second stream, then vapours from the second stream are used to heat up the third stream using a third heat exchanger. The second modification was towards lowering the high water waste production. The metrics provided in Fig. 6 showed a very high production wastewater (0.55 kg/kg-product). But a solution for the wastewater can be proposed in terms of installing a water treatment plant. By installing treatment, the wastewater being produced can be purified and recycled back into the process. As the water being produced is equal to the input requirement, therefore no makeup water would be required. For the purpose of this study, the water treatment has not been designed or simulated, but instead the cost of treating water was found to be 1.73 $/m³ (Oil and Gas Accountability Project, 2007; Saltworks, 2019), and this value was used for the TEA of the improved design, and for the sustainability analysis, it is assumed that all of the wastewater is treated and recycled back into the process.

3.2.2. Techno-economic analysis

For the improved design, the TEA was performed, and the comparison in Fig. 7 shows that the TCI for the improved design is higher, due to the addition of water treatment plant and the three flash tanks and heat exchangers, which are going to be used for the multiple effect evaporator. The TMC is decreased by 2 MMUSD/yr, because the high cost of water disposal and water purchasing has been replaced with a single cost of water treatment. Using water treatment, the treated water can be recycled and there is no need of buying any makeup water. Upon these modifications, the NPV was calculated to be higher, because for the same amount of sales, there are now a lower TMC. These results indicate that these modifications are capable of bringing an improvement in terms of economics.

3.2.3. Sustainability analysis

The two comparison metrics, Energy/Product and Waste/Product indicate the energy needed to produce and amount of waste produced per 1 kg of product respectively. The third metric indicates the amount of indirect CO₂ being emitted. As shown by Fig. 8, the energy requirement for the improved design was reduced by 22%, because by using the MEE strategy, only one-third of the water stream needs to be heated using utility, while the other are heated using the heat exchangers. The waste production was reduced by 80%. In the base case design, the 0.68 kg-waste/kg-product included 0.55 kg-wastewater/kg-product, but by adding a water treatment plant, all that wastewater is recycled and is no longer a waste stream. Therefore, in the improved design the total waste produced is 0.13 kg/kg. The remaining waste includes 0.03 kg/kg and 0.09 kg/kg of hazardous and non-hazardous waste respectively. These streams could not be recycled back into the process, therefore options for them other than disposal are limited. Finally, as the energy consumption has been reduced, the indirect CO₂ emission has also been reduced by 20%. Meanwhile, the overall reduction remained the same as 90% as the base case design.

4. Conclusions

On the basis of techno-economic analyses performed on the four scenarios, it can be concluded that the most profitable scenario is Scenario 3, the integration of BDF with bio-SA production. It is because the additional sales from bio-SA covers the new process’ operating expenses and the sales of BDF are not burdened to carry the entire profitability. This further supports the conclusion that the production of bio-SA from waste glycerol is more profitable rather than disregarding glycerol as waste stream. In the case of integration with MeOH, additional expenses are added onto the process without adding any additional revenue. This puts a constraint on the BDF sales, causing the minimum selling price to increase. From the sensitivity analysis, it is concluded that the variables to which the scenarios are the most sensitive is BDF price, WCO price and income tax.

In terms of sustainability, it can be concluded that Scenario 3 is the most optimum scenario. Even though the total waste production in Scenario 3 is higher (0.68 kg/kg-product) compared to Scenario 2 (0.36 kg/kg-product), 40% of the waste in Scenario 2 is hazardous waste, compared to 6% of Scenario 3. Meanwhile the 80% of waste production in Scenario 3 is wastewater. Secondly, in terms of indirect CO₂ emissions, Scenario 3 has 3 times less CO₂ emissions than Scenario 2, but double compared to Scenario 1. Furthermore, due to the CH₄ emission in Scenario 2, the CO₂ emission are 520 times more than Scenario 3. Therefore, the environmental impact in Scenario 3 is concluded to be less detrimental than Scenario 2 and 4.

Finally, for Scenario 3, different improvements such as multi-effect evaporator and wastewater treatment were implemented and investigated. For the TEA, the TCI for the improved design was higher due to the additional equipment, meanwhile the TMC decreased by 2 MMUSD/
yr. Upon these modifications, the NPV was calculated to be higher, because for the same number of sales, there are now much lower annual expenditures. These results conclude that the selected modifications favor the economics. Secondly, in terms of sustainability, the energy requirement for the improved design was reduced by 22% compared to the base case design, because by using the MEE strategy, utility expenditures. These results conclude that the selected modifications because for the same number of sales, there are now much lower annual expenditures. Meanwhile, the overall reduction remained the same at 90%. The consideration of geographical implications is subject to future investigations.

**List of abbreviations**

- WCI: Working Capital Investment
- WCO: Waste Cooking Oil
- WT: Wastewater Treatment
- MEE: Minimum Energy Expenditure
- TEA: Techno-economic Analysis

**Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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**Appendix A. Supplementary data**

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2021.130317.

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