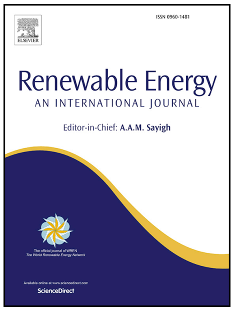

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Author Contributions

Danielle J. Nouwe Edou contributed to the process simulation, process modelling, generation of economic analysis data and editing of the manuscript. Jude Onwudili's contribution included project conceptualisation, funding acquisition, methodology design, supervision of process design and economic analysis, project administration and writing of the paper ((draft, review and final editing).

Process modelling and economic evaluation of biopropane production from aqueous butyric acid feedstock

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Abstract

Catalytic hydrothermal decarboxylation of biomass-derived butyric acid can produce renewable biopropane as a direct drop-in replacement fuel for liquefied petroleum gases. In this present study, experimental results from a batch reactor have been used to develop a hypothetical continuous process to deliver 20,000 tonnes/year of biopropane, as base-case capacity, from 10 wt% aqueous butyric acid. A combination of process synthesis and ASPEN Hysys simulation have been used to formulate a process flowsheet, after equipment selection. The flowsheet has been used to carry out economic analyses, which show that the minimum selling price of biopropane is 2.51/kg without selling the CO_2 co-product. However, with the incorporation of existing UK renewable energy incentives, the minimum selling price can reduce to $0.98/kg, which is cheaper than the current $1.25/kg selling price for fossil liquefied petroleum gases. Sensitivity analysis based on raw material costs and production capacities show profound influence on the minimum selling price, with strong potentials to making biopropane competitive without incentivisation, whereas the influence of selling CO_2 is marginal. While this biopropane technology appears promising, it still requires more detailed technical and process data, life-cycle analysis and detail economic costings for testing at a pilot-scale prior to commercial exploitation.

Keywords: biopropane, hydrothermal decarboxylation, butyric acid, processing synthesis, process modelling, economic analysis

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1.0 Introduction

Biomass-derived liquefied petroleum gases (bio-LPG) is a low-carbon direct drop-in biofuel, which has the same chemical and physical characteristics as the main components (propane and butane) of fossil LPG. According to the World Liquefied Petroleum Gases Association (WLPGA) and Liquid Gas UK, (LGUK), there are over 2 million off-grid UK homes that can be supplied with sustainable low-carbon fuels and energy for heating [1, 2]. Both the WLPGA and LGUK have proposed bio-LPG as a potential solution for off-grid heating to reduce carbon emissions [1, 2]. Studies by UK Government Department for Business Energy and industrial strategy [3] and the renowned bioenergy consultant, NNFCC [4], agree that bio-LPG has the potential to satisfy UK's residual gas needs, especially for rural off-grid heating. Therefore, commercial production of bio-LPG has become a topical research interest, driven by decarbonisation targets and the desire to deliver this product at a large scale and at competitive prices.

Currently, the main process for making bio-LPG is via hydroprocessing of vegetable oils and fats (HVO) to produce green liquid fuels such as gasoline, kerosene and diesel as the target products. In this process, biopropane is obtained as a by-product at a yield of 5 – 8% [5]. As a result, the current commercial production plant capacity for HVO biopropane is limited to around 40,000 tonnes per year and the global capacity is around 220,000 tonnes per year [6]. Therefore, to meet growing demand for bio-LPG, dedicated plants producing larger quantities of biopropane or biobutane (or both) as main products will be required. This is especially so, given that the markets for green liquid hydrocarbons, which currently drive HVO bio-LPG, are destined to face a downward trend with the impending ban on internal combustion engines (ICE) operating on these types of liquid fuels [7].

One potential process route to make biopropane as a major commercial product is via the catalytic hydrothermal decarboxylation of butyric acid [8, 9]. Decarboxylation of butyric acids would theoretically produce 50 wt% of propane, which is significantly higher than the yield of propane obtained from other biomass feedstocks reported in literature. This, therefore, provides a good starting point for its commercial exploitation. Butyric acid can be produced from biomass via fermentation processes [10, 11]. Recently, a novel fermentation process using *Clostridium tyrobutyricum* immobilized in a fibrous bed bioreactor has been reported to produce a butyric acid concentration of up to 86.9 g/L, which is the highest butyric acid concentration ever produced [11]. However, the low concentrations of butyric acid obtained from these fermentation processes will require expensive water removal and purification to obtain the dry product [12].

Hydrothermal decarboxylation, which occurs in water medium, does not require extensive dewatering and therefore is suited to use aqueous butyric acid solutions as a sustainable feedstock, thereby potentially reducing raw material costs compared to the purified and dried feedstock. The butyric acid route must also contribute to greenhouse gas (GHG) reduction at the same level or better than other existing or developing bio-LPG production routes. Studies, based on the HVO route, have shown that the carbon intensity of bio-LPG can be around 80% lower than oil [2], and for the UK, switching from fossil LPG to bio-LPG can potentially lead to around 78% reduction in GHG emissions [4]. In addition, bio-LPG, in general, has the potential to improve air quality, giving that its combustion emits 27% less nitrogen oxides (NOx) and 43% less particulate matter compared to oil [2]. Although, detailed life-cycle analysis will be needed confirm any GHG emission reduction that can be obtained from this butyric acid route, it has the potential to deliver similar GHG savings as the other bio-LPG routes. For instance, at the conversion technology stage, the HVO route uses large amounts of hydrogen to remove oxygen from vegetable oils as water to make propane and green hydrocarbon liquid fuels. Hydrogenation avoids direct CO_2 release during the HVO process. In contrast, the butyric acid route does not require hydrogen to make propane but produces CO_2, which can also be converted to methane or methanol using similar or lower amounts of hydrogen compared to the HVO route. Therefore, GHG reductions from the two processes may be technically similar, provided the hydrogen feedstock required is obtained from the same sustainable source.

Recent publications on hydrothermal conversion of butyric acid to propane have been carried out in laboratory-scale batch or flow reactors [8, 9]. Using, platinum-based catalysts, the reaction to convert butyric acid is efficient at around 300 °C – 350 °C and pressure of up to 21,000 kPa [9]. Results from laboratory tests can be used to begin to synthesise a complete process from feedstock to product using chemical and process engineering tools and skills. As part of chemical process design, preliminary techno-economic analyses can be carried out to determine the potential economic viability of a process route for potential commercial exploitation. This is the aim of this present novel study, which is the first to carry out any process modelling and techno-economic analysis of biopropane production from aqueous butyric acid. The results of this preliminary work would provide some indication of the technological and economic viability of the butyric acid process route, which can contribute to future process development.

1.1 Background Information

The present study is based on experimental data from a laboratory-scale batch reactor that gave the highest conversion of butyric acid (97.8%) and the highest biopropane yield of 46.4% and a biopropane hydrocarbon selectivity of 96% [9]. In the experiment, 1.0 g of butyric acid was dissolved in 9.0 g of water to make at 10 wt% solution, which was found to be the optimum butyric acid concentration to give the highest conversion efficiency [9]. In comparison, butyric acid concentrations from fermentation processes are typically between 3 - 9 wt% in aqueous solution [10 -11, 13]. Higher concentrations of up to 30 wt% can be obtained via extractive fermentations [13]. Therefore, the feedstock concentration of 10 wt% can be achieved by reducing the water content in fermentation broths, or by diluting the concentrated product from extractive fermentation. These measures can deliver energy savings compared to using the dried solid product as feedstock. The 10 wt% solution was reacted at 300 °C and autogenic pressure of 12,000 kPa in the presence of 1.0 g of 5.0 wt% Pt/C catalyst (nominal Pt content of 0.05 g); which gave a butyric acid to active metal catalyst mass ratio of 20:1 [9].

Using the experimental data, the main chemical reactions leading to the formation of the products observed during the hydrothermal decarboxylation of aqueous butyric acid were derived.

The main reaction was the conversion (97.8%) of butyric acid via decarboxylation in the presence of Pt/C catalyst according to Equation 1.

$$C_4H_8O_2 \text{ -> } C_3H_8 + CO_2 \qquad (1)$$

Complete decarboxylation of butyric acid would yield a 1:1 molar ratio of propane and carbon dioxide but results under the reaction conditions showed that the molar concentration of CO_2 in the gas was slightly higher than that of propane [9]. This indicated that propane was being consumed by other reactions and in addition, the gas product was found to contain some hydrogen, methane, and ethane. Based on detailed analysis of the gas products, it was inferred that the formation of these gases could be via aqueous reforming of butyric acid (Equation 2) and hydrogenolysis of the propane product (Equation 3). Experimental data showed that the concentrations of both CO_2 and methane increased while that of propane decreased with increasing reaction temperature, making these reaction mechanisms plausible [9]. Therefore, based on the total yield of CO_2 [9], it was estimated that about 2.22% of the butyric acid was reformed via Equation 2.

$$C_4H_8O_2 + 6H_2O \dashrightarrow 4CO_2 + 10H_2 \qquad (2)$$

The hydrogen produced from the reforming reaction could cause the hydrogenolysis of the propane product. Therefore, using the yields of methane and ethane obtained under the reaction conditions of 300 °C and 12000 kPa [9], the amount of propane converted via hydrogenolysis (Equation 3) was estimated to be about 1%.

$$16C_3H_8 + 11H_2 \dashrightarrow 6CH_4 + 21C_2H_6 \qquad (3)$$

2.0 Methodology

2.1. Design basis

The base case design basis is the production of 20,000 tonnes/year of biopropane from 10 wt% aqueous butyric acid from biomass. However, the production of butyric acid from biomass via fermentation has not been considered within the scope of this study. Instead, the cost of producing biomass-derived butyric acid has been found in peer-reviewed literature and used [14]. Therefore, the starting point is the input of 10 wt% aqueous butyric acid feedstock into the process and the end point is the output of 99.9% biopropane product. The experimental data obtained from the reaction at 300 °C and 12000 kPa [9] (explained above) were used for the modelling of a continuous process plant on ASPEN

Hysys. Thereafter, the biopropane production capacity is scaled up to 40,000 tonnes/y, 60,000 tonnes/y, 80,000 tonnes/y and 100,000 tonnes/y, respectively, for comparative economic analyses.

2.2 Process synthesis and simulation

The entire process for the production of biopropane from 10 wt% aqueous butyric acid has been designed from scratch by a combination of process syntheses and process simulation. The unit operations involved in the process were identified, followed by the selection of appropriate pieces of equipment to achieve the design intent. For process simulation, the Peng Robinson-Strygek-Vera (PRSV) fluid package on ASPEN Hysys has been used to calculate the physical properties of conventional substances during the conversion of aqueous butyric acid into biopropane and CO_2.

2.3 Process flowsheet

The main butyric acid conversion is a relatively simple process, based on decarboxylation to yield biopropane and CO_2 as the main products. Thereafter, downstream separation units are applied to obtain the biopropane product at the required specification. The ASPEN Hysys simulated process flowsheet resulting from this present study is provided in the *Supplementary Information* (Figure SI1). The flowsheet focuses on the main process from the butyric acid feed up to the amine absorption system, where the mixture of gases containing biopropane is obtained. Using the simulated flowsheet, a process flow diagram has been developed with AutoCAD P&ID and presented in Figure 1. The feed storage and mixing tanks as well as the biopropane recovery unit have been excluded process flow diagram but have been used in the economic analyses.

With the reactor operating at 12000 kPa, a pump (P-101) operating at 120 bar (12000 kPa) is used to transfer the required aqueous butyric acid feed, initially held in a bunded mixing tank at 20 °C and 100 kPa. The tanks are assumed to be made of aluminium, which is known from Corrosion Charts [15] to be resistant to 10 wt% acetic acid solutions for temperatures up to 60 °C, and so could be ideal for butyric acid solution of similar concentration. The feed is then pumped from the mixing tank through a pre-heater (HE-101), where it is pre-heated up to 80 °C, before it is passed through another a vapouriser (HE-102) operating at 180 °C (normal boiling point of butyric acid is 163.5 °C).

The high-pressure pre-heater and vapouriser are assumed to be of shell and tube type, with the aqueous butyric acid feed passing through the tubes made of stainless steel [16]. The vaporised feedstock mixture then enters a fixed bed reactor (R-101), where the conversion of butyric acid (97.8%) via the main decarboxylation reaction occurs at 300 °C and 12000 kPa. A conversion reactor has been selected by ASPEN Hysys, but this can be improved in future as more relevant experimental data become available. High-grade stainless steel, such as Alloy 20, for high temperature dilute acid environments has been selected as material for the reactor [16].

For energy integration, the outlet stream of the reactor is used as the heating fluid for the feed entering both the vapouriser and pre-heater before being further passed through a cooler (HE-103), cooling the product stream to 87 °C. All downstream equipment after the reactor is assumed to be constructed with carbon steel. The cooled product stream then enters a gas-liquid separator (V-101) where, based on the ASPEN Hysys model, 99.9 wt%

water is condensed and removed along with about 13.7 wt% of the CO_2, which dissolves in the condensed water under the process conditions. The remaining mixture consisting mostly of gaseous propane, carbon dioxide and traces of ethane, methane and water vapour is then transferred to a decompressor (C-101) operating at 6190 kPa before entering the amine absorber (V-102).

In the absorber, the vapour mixture encounters 227.53 kmol/h of monoethanolamine (MEA) solution consisting of a 1:1 molar ratio of water and MEA [17]. It is assumed that the high-pressure amine absorber [18] removes all the CO_2 and remaining water vapour from the process stream, leaving propane, hydrogen, methane, and ethane in the gas-phase product (biopropane-rich product). The rich amine stream is then firstly heated to 120 °C by HE-104 before entering the amine regeneration unit (V-103), where CO_2 is removed at the top, and the lean amine used as the heating fluid for HE-104 and cooled to 60.2 °C before being recycled to the amine absorber. While it is not shown in Figure 1, the biopropane-rich product gas is assumed to be sent into a straight refrigeration unit [19], where liquid biopropane is recovered at a purity of 99.9%. The straight refrigeration process for biopropane recovery has been chosen as it is cheaper than lean oil absorption and cryogenic processes [19] for LPG production.

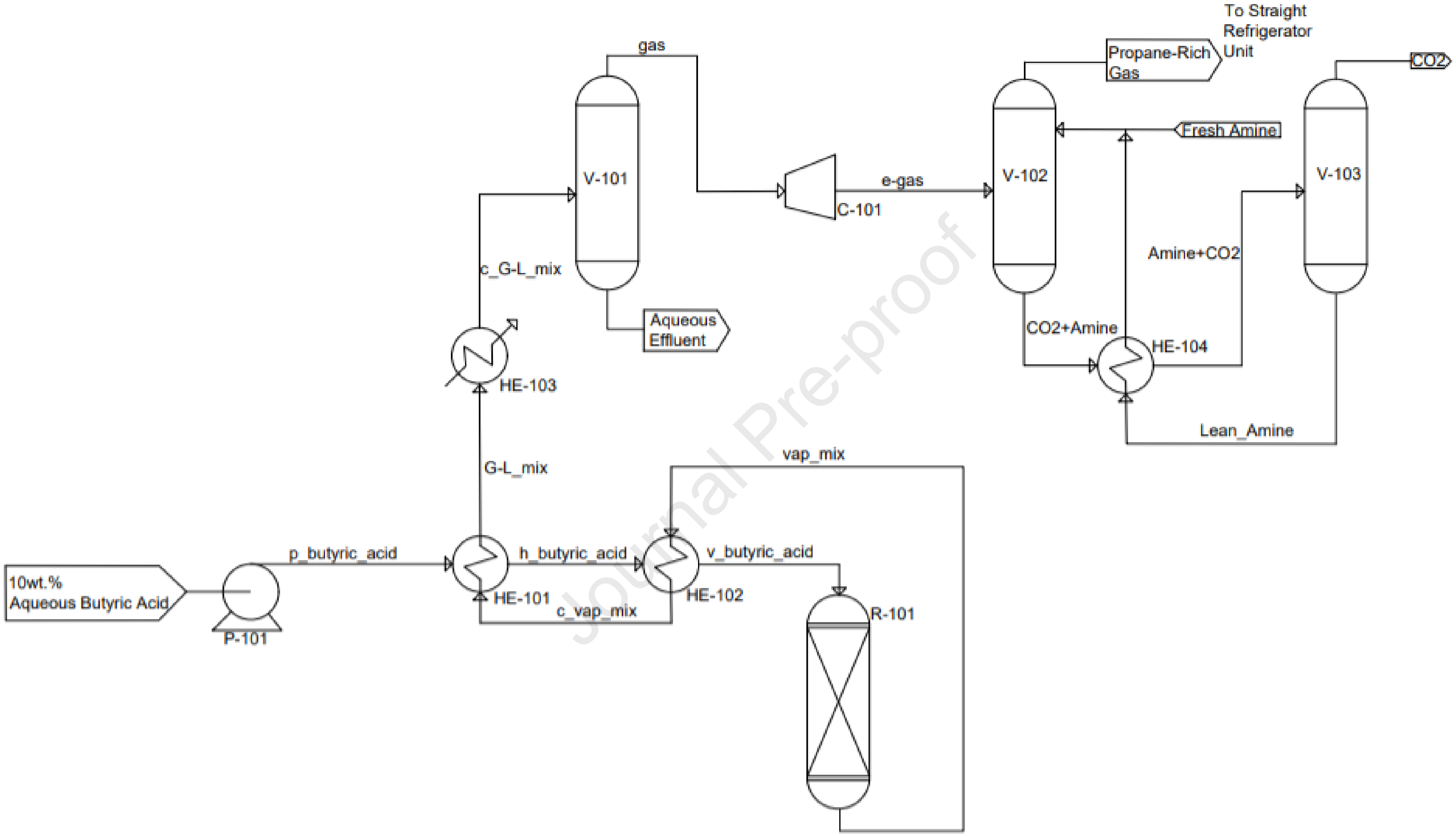

Figure 1: Simulated process flow diagram for biopropane production from catalytic decarboxylation of butyric acid

2.4 Economic assessment

The basic assumptions used for the economic analyses of the base case scenario are presented in Table 1. Interest rate on capital borrow has been taken as 7.5% due to the process

Table 1: Assumed economic parameters

Economic parameter	Value	Unit	Ref
Plant life	20	Year	[20]
Annual operating hours	7920	hours/year	-
Exchange rate (£ to $)	1.36		[21]
Income tax charge	30	%	[20][22]
Interest rate	7.5	%	[22]
Internal rate of return (IRR)	10	%	[22]
Catalyst costs (% of total equipment cost)	10	%	[24][25]
Biomass-derived butyric acid cost price	580	$/tonne	[14]
Process water cost	3.4	$/tonne	[26]
Amine raw material cost (including disposal)	6.74	$/tonne	[27]
Operator salary	35,000	$/year	[28]

being high risk (new technology for a product with an existing market) [22, 29]. In addition, raw material costs and product selling prices have been assumed to be constant over the 20-year period, thereby neglecting fluctuations in costs due to general inflation or processing demands when calculating the Net Present Value [29].

2.4.1. Capital costs

Using the obtained process flow diagram, mass balances and energy balances, the sizing of each equipment has been determined based on the mass flow rates, densities and assumed residence times [15, 29]. The cost of the main reactor fixed bed has been estimated using the ASPEN Process Economic Analyser software. For the heat exchangers (HX), (including pre-heater and vapouriser), their types, sizes and surface areas were determined from thermal design calculations [29] and these were used to calculate their costs. All heat exchangers were assumed to be of fixed-tube sheet shell and tube type, constructed with stainless steel; hence, using a type factor of 0.8 and pressure factor of 1.5. The purchase costs of the heat exchangers were obtained according to Equation 4 [29].

Purchased cost of HX = Bare cost x Type factor x Pressure factor (4)

The downstream biopropane recovery unit is assumed to be similar to the one used for the recovery of fossil LPG from refinery off-gases [19]. Therefore, in this study, the inside battery limit (ISBL) cost of the biopropane recovery unit has been obtained using the Historic Cost Data method [29] based on a value found in literature for a plant operating at an LPG flow rate of 17,545 kg/h [19], according to Equation 5.

$$ISBL\ capital\ cost\ of\ new\ biopropane\ recovery\ unit\ = Cost_{old}\left(\frac{Capacity_{new}}{Capacity_{old}}\right)^{n} \tag{5}$$

Where, the average value of n = 0.6 for the chemical industry has been used.

In addition, the costing of other pieces of equipment such as gas-liquid separators, pumps, compressors, amine scrubbers and storage tanks have been obtained from published price data from relevant Chemical Engineering Economics literature [30-32]. To obtain prices for 2021 (year of study), the Oil and Gas Field Machinery and Equipment Manufacturing Cost Index [33], published in 1989 has been used according to Equation 6.

$$C = C_i \times \frac{Index_{2021}}{Index_{1989}} \times f \quad (6)$$

Where, C = present cost of equipment, C_i = historical cost of equipment, f = factor accounting for construction material type and insulation (f = 1.30 for stainless steel).

Using the total equipment cost, a detailed factorial method has been used to estimate the capital costs of other components of the plant on US Gulf Coast Basis [29, 30]. These components included pipe works, buildings, land, instrumentation, installation and construction labour and supervisor, construction costs, electrical, site, construction costs, buildings and structures, services, and engineering. Following this, the realistic capital cost of the UK-based plant has been obtained by applying location factors for USA and the UK [29].

2.4.2 Operating costs

In process plants, operating costs are typically made up of raw materials costs, labour costs and other costs. The cost of feedstock (10 wt% butyric acid solution) has been estimated from the purchase prices of components shown in Table 1, for dried biomass-derived butyric acid [14] and process water [26] and raw material requirements for the amine absorber [27]. In chemical plants, the cost of catalysts can vary between 3% and 10% of total equipment costs [24, 25]. Considering that the platinum is an expensive metal, the cost of 5 wt% Pt/C catalyst has been estimated using the upper limit of 10% of the purchased cost of equipment. The labour cost has been obtained via the capacity-labour chart [29] and $35,000 per annum used as the likely wage of UK-based plant operators [28]. Other components of the operating costs were calculated using typical factors [29]

2.4.3. Economic performance analysis

The economic evaluation of the aqueous butyric acid route to biopropane has been carried out in this study to determine net cash flow (NCF), breakeven point, payback period, net present value (NPV) and return on investment (ROI). The values of these parameters have been obtained based on their usual formulae [29]. For the economic analysis, the minimum selling price of the biopropane product under different system and market scenarios are obtained and compared to the current selling price of fossil LPG in the transport sector as the reference product. For each analysis, the minimum selling price of biopropane has been obtained with the expectation that the breakeven point (when revenues equal total production costs) would occur when the operating capacity of the plant is below 70%, to ensure profitability while accommodating fluctuations in operational contingencies and market demands [29]. To do this, the selling price of biopropane has been varied within a given reasonable range to obtain values for net NCF, NPVs, payback periods and ROI at the fixed internal rate of return (IRR) assumed in this work (10%) [22]. The minimum selling price of biopropane results from the biopropane price value that gives an NPV of zero for the plant [34].

2.4.4 Sensitivity analysis

The scale-up of the plant to higher biopropane production capacities from 20,000 tonnes/year to up to 100,000 tonnes/year and potential changes in raw material costs between –80% and +60% of the base case value have been used for sensitivity analyses. In addition, since the process involved the separation and removal of CO_2 using an amine absorber, this can be captured, stored, and sold (or not sold) for income also. Therefore, the selling price of CO_2 (between \$0 and \$200 per tonne) has been considered in sensitivity analyses. The influence of these parameters on the minimum selling price of biopropane, payback periods and return on investment at 10% IRR have been determined.

3.0 Results and Discussion

3.1 Mass balance and energy balances

The Blackbox mass balances for the plant designed to produce 20,000 tonnes of biopropane per year (i.e., 2,525.25 kg/h) is presented in Table 2. Data from this table can be very useful for calculating some elements of operating costs during economic analyses. For instance, the large flow rate of feedstock (52,799.54 kg/h) would require large capacity chemical storage tanks made with appropriate material to provide enough quantity of feedstock for continuous operation. Alternatively, the aqueous feed can be made up on site using solid butyric acid and process water to reduce cost of feed storage equipment (tanks).

Table 2: Blackbox mass balances based on ASPEN Hysys simulation

	IN (kg/h)		OUT (kg/h)	
Components	Reactor	Amine Absorber	Gas-Liquid Separator	Amine Absorber
Butyric acid	5,279.954			
H_2O	47,519.583	1,024.747	47,473.179	1,047.185
MEA Solution		3,474.615		3,474.615
Propane				2,525.253
CO_2			361.542	2,275.810
CH_4				21.333
C_2H_6				119.953
Total	52,799.537	4,499.362	47,834.733	9,464.150
Grand Total	57,298.899		57,298.883	

The main energy requirements of the 20,000 tonnes of biopropane per year process plant is also shown in Table 3. Overall, producing this amount of biopropane from this butyric acid route required net energy input overall, at the rate of 4.3 MW. The process has been designed as shown in Figure 1 to use the reactor product stream for the pre-heating and vaporisation of the feed stream, thereby helping to reduce the overall energy requirement.

Table 3. Overall energy balance around main equipment

Component	Heat Flow (kJ/h)
Pump	+834,610.59
Pre-heater	-
Vapouriser	-
Reactor	+41,938,472.35
Reactor Product Cooler	- 27,603,634.93
Gas Product Cooler/decompressor	-64,973.40
Amine Absorber	+287,688.67
Total (kJ/h)	+15,392,163.27
Total (kW)	+4275.60

3.2. Results of economic analyses

3.2.1 Capital costs

The calculated costs of all major pieces of equipment are presented in Table 4 based on Aspen Economic Analyser, thermal design and literature [30-32]. The solid butyric acid is stored in a 50 m^3 glass-lined (to prevent corrosion) carbon steel tank, operating at atmospheric pressure and can hold up to one month requirement of this feedstock. Hence, the 10 wt% aqueous butyric acid is made up continuously and used on site in another 50 m^3 glass-lined carbon steel feed mixing tank. This arrangement has been found to be over 7 times cheaper than using large capacity tanks to store 5000 m^3 of ready-made 10 wt% aqueous butyric acid solution [30]. The cost of the biopropane recovery unit has accounted for more than half of the total equipment cost (65.3%). This has been estimated on ISBL basis, with the whole unit comprising of several pieces of equipment to recover 99.9% of biopropane from the biopropane-rich gas products exiting the amine absorber. The capital costs of other items have been estimated using the typical lang factors [29]. and presented in Table 3. With these a total plant cost (TPC) of about $21.8 million has been obtained and with inclusion of other capital cost elements, such as contractors' fees, contingency fees and working capital (charged typically at 15% of TPC), the total capital cost of the plant in the UK has been obtained as $26 million, after applying the location factor (1.11) for UK. The scrap value has been estimated to be about $8.91 million, comprising of 50% of the cost of Buildings and Structures and 25% of other eligible capital cost elements, including process equipment.

Table 4: Estimated total capital costs for the production of 20,000 tonnes/y biopropane from aqueous butyric acid

Main Equipment	Cost ($)
Feed storage/mixing tanks (2 x 50 m^3)	156,385.46
Pump	221,700.00
Pre-heater	282,681.23
Vaporiser	512,255.92
Fixed Bed Reactor	249,900.00
Heat Exchanger (E-100)	75,600.00
G-L Separator	119,300.00
Decompressor	93,000.00
Amine Absorber	33,100.00
Heat Exchanger (E-101)	11,300.00
Amine Regenerator	33,100.00
Heat Exchanger (E-102)	11,000.00
Biopropane recovery unit	3,385,802.62
Total Purchased Cost of Equipment	5,185,125.22

Other Capital Cost Items	Lang Factor	Purchased Cost ($)
Purchased Equipment	1	5,185,125.22
Piping & Ductwork	0.47	2,437,008.85
Instrumentation	0.18	933,322.54
Building & Structures	0.66	3,422,182.65
Installation labour & supervision	0.11	570,363.77
Electrical	0.18	933,322.54
Site	0.1	518,512.52
Services	0.7	3,629,587.66
Land	0.06	311,107.51
Construction Costs	0.41	2,125,901.34
Engineering	0.33	1,711,091.32
SUBTOTAL (TOTAL PLANT COST)	4.2	21,777,525.93
Contractors' fee @ 5% TPC	0.21	1,088,876.30
Contingency @ 10% TPC	0.42	2,177,752.59
Total Fixed Capital Cost (FCI)	4.83	25,044,154.82
Working Capital (15% $FCI)		3,756,623.22
Capital Cost in USA ($)		28,800,778.04
Scrap Value ($)		8,911,285.83
Capital Cost in United Kingdom ($)		25,946,646.89

3.2.2 Operating costs

Table 5 presents the operating costs obtained for the biopropane production plant based on a capacity of 20,000 tonnes per year.

Table 5: Components of the estimated operating costs to produce 20,000 tonnes of biopropane per year

Items	Cost ($)	Classification
Raw materials	27,181,203.19	Variable Costs
Catalysts	2,594,664.69	
Labour	733,055.40	
Supervision and Management	146,611.08	
Utilities	2,039,764.10	
Maintenance	2,071,183.14	
Overheads	2,657,627.46	
Depreciation	1,704,454.13	Fixed Costs
Insurance	345,197.19	
Interest on capital (7.5%)	2,682,275.47	
Rates and Taxes	1,035,591.57	
Rent	517,795.79	
Administration	146,611.08	General Cost
Distribution	4,510,635.67	
Research and Development	2,050,288.94	
Total	50,416,958.90	

The total raw material costs have been calculated based on individual costs of biomass-derived butyric acid, process water and amine absorber [14, 26-27]. The butyric acid feedstock requirement for one operating year comes to $24.25 million based on$580/tonne [14]. This value for butyric acid should be seen as already high, considering that water removal via distillation is the major cost item in order to obtain the dried product. It is also possible that, butyric acid costs would fall with investments into large-scale biotechnological processes to produce the aqueous feedstock, with the inherent advantages of economies of scale. Process water will cost about $1.28 million per year at a cost of $3.4/tonne [26], while the raw material cost for the amine absorption system is about $1.65 million per year.

Using these values found in literature, a total raw material cost of $27.18 million has been obtained for the base case scenario of 20,000 tonnes/year. The decarboxylation of butyric acid to obtain biopropane for this study is based on the use of expensive platinum-based catalysts. The costs of catalysts are typically estimated to be between 3% and 10% of total equipment costs [24-25]. With Pt being an expensive metal, the catalyst cost has been estimated at the upper limit of 10% of the total equipment costs, thereby giving a catalyst cost of $2.59 million per year. The labour cost has been estimated after obtaining the number of operating hours from the plant capacity chart [29]. As there are four main processing steps, 2 shift patterns and 3 shift positions, with 3 operators per shift, giving a total 18 operators per day have been estimated. The total labour cost per year is $733,055.

3.2.3 Biopropane production costs

In this study, all capital investment funds were borrowed at an annual interest rate of 7.5%, with depreciation charged over the 20-year life span assumed for the project. Under this scenario, the total production cost of biopropane is $2,337.90/tonne. This value is slightly higher than the unit cost of HVO biopropane, estimated to be $2,312.51/tonne in 2013/2014 [35]. Considering only the cash cost (i.e., excluding cost of capital and depreciation), the cost of making biopropane from butyric acid reduces to $2,125.62/tonne, which is still higher than the equivalent cash cost of HVO biopropane of $1,960/tonne [35]. However, the HVO biopropane is produced as a by-product at plants with larger production capacities and therefore may enjoy the advantages of economies of scale.

3.3 Biopropane Minimum Selling Price

Figure 2 presents the plots of the return on investment (ROI), payback periods and net present values, used to determine the minimum selling price of biopropane obtained from the butyric acid process. The minimum selling price is obtained when the NPV becomes zero, without the CO_2 co-product being sold. Under this scenario, biopropane would sell for \$2.51/kg, an ROI of 7.56% and a payback perio of 8.93 years (Figure 3).

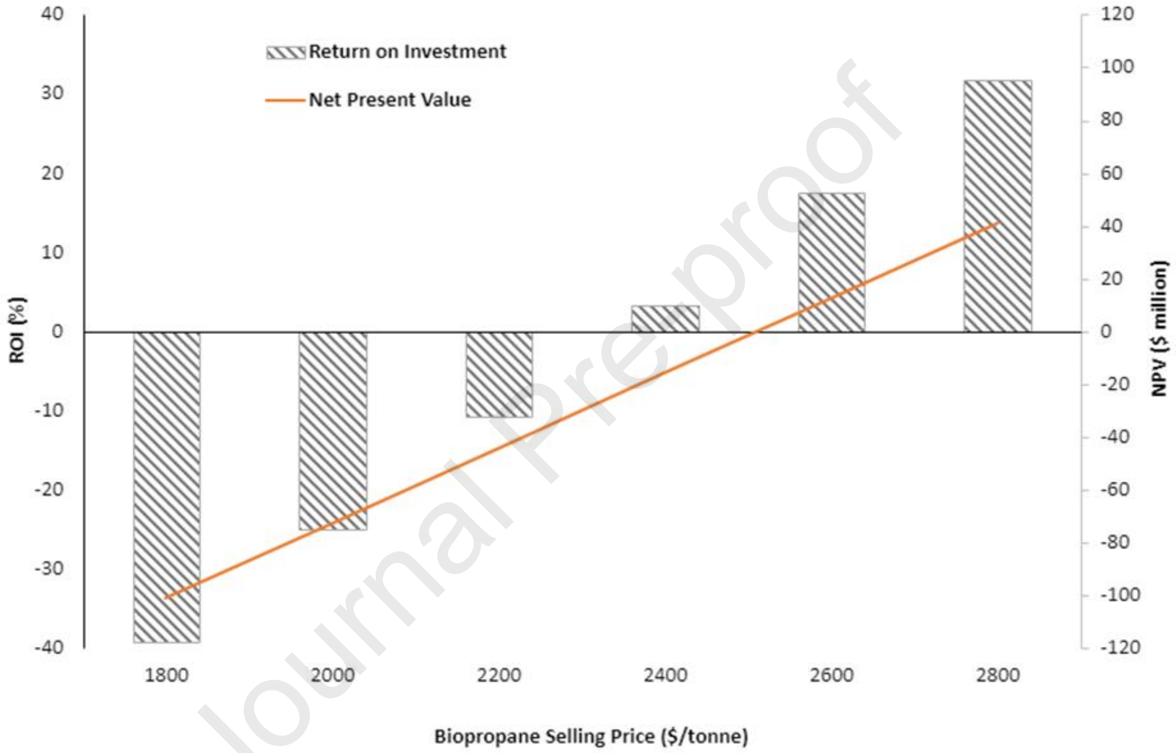

Figure 2: Determination of biopropane minimum selling price at 20,000 tonnes per year production capacity

The cost of biofuels is often incentivised to encourage increased uptake and make them more affordable. Different schemes exist in different domains and in the UK, the applicable scheme is the Renewable Tranport Fuel Certificates (RTFCs) and green house gas (GHG) credits [4]. The combined general RTFC and GHG credit incomes (GRTFCs) for supplying bio-LPG in the transport market is around \$1.53/kg [4], therefore this can bring the selling price down to a more affordable \$0.98/kg.

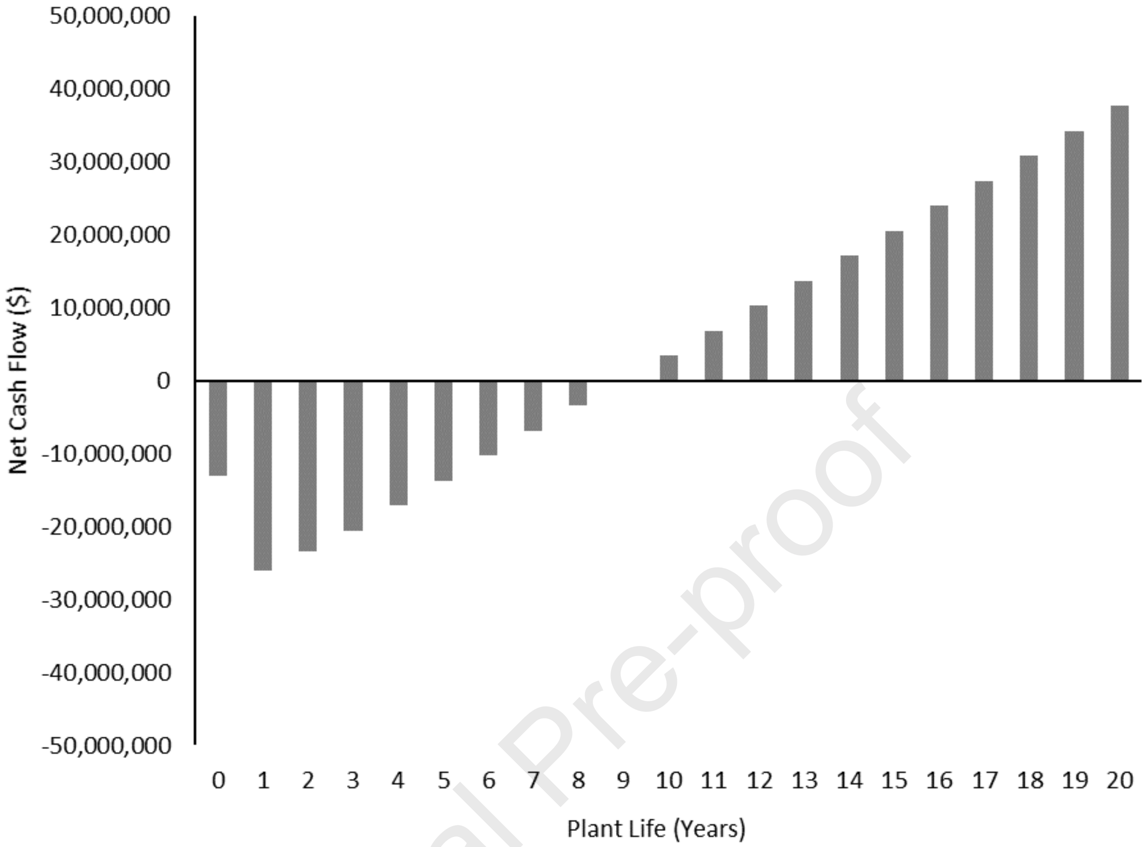

Figure 3: Determination of payback period for 20,000 tonnes/year biopropane capacity

According to the WLPGA [3], the fuel cost of a bio-LPG boiler is estimated to be \$0.1014/kWh, and using the average calorific value of propane (48.5 MJ/kg), this translates to purchase price of \$1.37/kg at current exchange rates, which is higher than the \$0.98kg obtained above. Considering that most bio-LPG currently being sold at the moment comes from the HVO route, the incentivised \$0.98/kg selling price of biopropane from butyric acid route may compete favourably, even without income from the CO_2 co-product. The reference product for comparison is commercial fossil LPG, which currently sells at a global average price of \$1.25/kg as transport fuel [4]. However, this butyric acid technology can produce pure biopropane and, the refill price of non-transport (domestic or commerical) fossil propane, ranges from \$4.11/kg to \$6.58/kg depending on cylinder size [36]. The pure biopropane produced via this technology can be sold at a premium for domestic and commerical applications, with higher profit margins.

In comparison, other techno-economic studies have obtained biopropane selling prices of \$1.05/kg, \$0.75/kg and \$0.95/kg for HVO process, biomass gasification and glycerol route, respectively [35, 37]. On the surface, it would look like these other technologies could deliver biopropane at similar price as the butyric acid route, however, the HVO and gasification routes rely on the sales of much larger product volumes of hydrocarbon fuels (green diesel and gasoline) to obtain the low biopropane selling prices [35]. Although, the biopropane minimum selling price has been obtained without selling the CO_2 co-product, the recommended CO_2 price range to support the scale-up of CO_2 utilisation is between \$40/tonne to \$80/tonne [38], which is about 360 times lower than the selling price of green diesel or gasoline (commercial diesel and gasoline each currently sells above \$2000/tonne). So, the influence of income from CO_2 sales may not be significant and this is presented in Sub-section 3.4.2.

In addition, the production volume of green diesel and gasoline are certianly much larger than biopropane from either biomass gasification and HVO, so the impacts they can make to the eventual selling price of biopropane is hugely significant. However, with the impending ban on internal combustion engines (ICEs), the incentives to make green diesel and gasoline would disappear, which will lead to a potentially high true selling price of biopropane from HVO and gasification. Also, the glycerol route may appear competitve but depends heavily on the cost of glycerol and hydrogen. The current cheap price of glycerol is mostly driven by its over-production as a by-product of the transesterification process to make biodiesel.

Therefore, scaling down biodiesel production due to the imminent ban of ICEs, will reduce availability of glycerol and potentially lead to price increase. Hence, it is expected that if biopropane is the main product being sold from these other routes, then its selling price would most probably be even higher than biopropane obtained from the present butyric acid route. Therefore realistically, it appears that biopropane from the butyric acid route can stand in good competition with the alternative technologies. In addition, the production capacity used in this study (20,000 tonnes/year) is much lower than the average of between 40,000 and 50,000 tonnes/year recommended for HVO biopropane [4] and even much larger capacities for biomass gasification route. Therefore, giving the possible economy of scale that comes with larger volume production, the butyric acid route could potentially deliver lower minimum selling prices at higher production capacities as presented in Sub-section 3.4.3.

3.4 Sensitivity analysis

3.4.1 Effect of CO_2 sales income on minimum selling price of biopropane

Carbon dioxide is the co-product of the decarboxylation of butyric acid to make biopropane. Theoretically, complete decarboxylation of butyric acid would produce a 1:1 molar ratio of propane and CO_2. Interestingly, both compounds have the similar molecular mass of about 44 g/mol, so that they would also have a 1:1 mass ratio in the product gas. However, it must be noted that while CO_2 production from this butyric acid process route is high, it is similar to the yield of CO_2 from the well-established and accepted fermentation of glucose to produce bioethanol fuel ($C_6H_{12}O_6 \rightarrow C_2H_5OH + 2CO_2$). For example, 0.96 kg of CO_2 is generated for 1 kg of bioethanol, while butyric acid decarboxylation generates 1 kg of CO_2 per kg of biopropane produced. It is therefore important to consider how the CO_2 should be handled, as it can have both environmental (if emitted) or economic implications (if sold for income) on the process. In this butyric acid process route, CO_2 can be recovered from the amine absorber and can be sold, without significant upstream costs. CO_2 utilisation is beginning to gain serious attention, with possible markets in the food and beverages industry, algae cultivation and in the chemical industry for chemical transformation to make high-value products. Hence, income from CO_2 may help reduce the selling price of biopropane produced from this technology. Figure 4 shows the effect of varying the CO_2 selling price from \$0/tonne to \$200/tonne, using a fixed IRR value of 10%. A recent techno-economic analysis on hydrogen production from biomass gasification indicated that CO_2 selling price would need to be above \$120/tonne to match the reference selling price of hydrogen from steam methane reforming [34]. In Figure 4, the ROI and payback periods remain consistent with average values of 7.20% and 8.95 years, respectively.

Increase in CO_2 selling price leads to consistent but marginal decrease in biopropane selling price, such that at a CO_2 selling price of \$200/tonne, biopropane selling price decreased by 7.75% (\$2.32/kg) compared to when not selling CO_2 (\$2.52/kg). Also, selling CO_2 around \$60/tonne [38] reduces the minimum selling price of biopropane by only 2.38%. Therefore, Figure 4 shows that the minimum selling prices of biopropane (without incorporating incentives) are still nearly double the reference LPG (for transport) selling price of \$1.25/kg, when CO_2 is sold within the given price range. Indeed, CO_2 would need to be sold at around \$1320/tonne, to bring the minimum selling price of biopropane down to \$1.25/kg. The prospect of achieving such high price for CO_2 is currently remote. However, incorporating the additional income from RTFCs and of about \$1.53/kg for bio-LPG [4], would deliver the product at \$0.985/kg even without selling CO_2 and \$0.79/kg at CO_2 maximum selling price of

CO_2 of \$200/tonne used in this study. Therefore, incentivisation is required to make biopropane from this butyric acid route cheaper than or cost competitive to the reference transport fossil LPG.

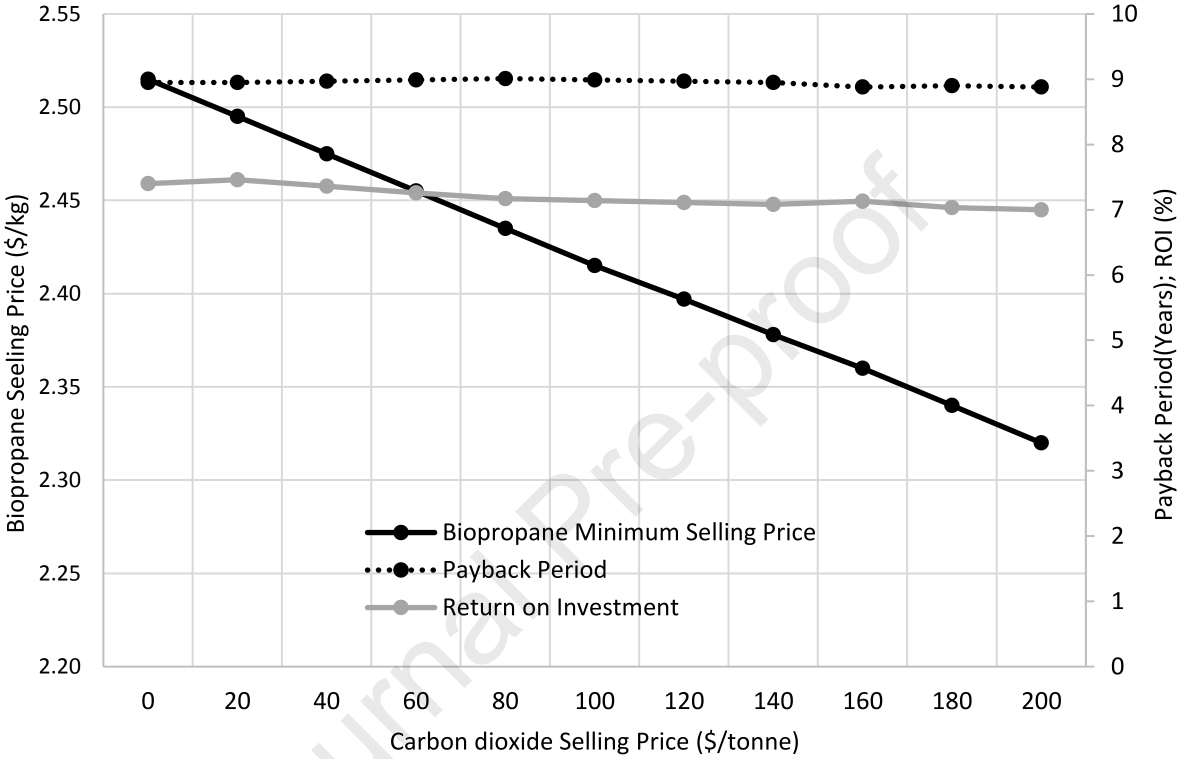

Figure 4: Influence of CO_2 selling price on the minimum selling price of biopropane at 20,000 tonnes/year production capacity

3.4.2 Effect of raw material costs on minimum selling price of biopropane

For the base case scenario (20,000 tonnes/year), the raw material cost has been calculated to be \$27.18 million. However, since aqueous butyric acid could be obtained and used from fermentation with minimal separation steps, the feedstock cost has the potential to be reviewed downwards. It could also go upwards due to economic uncertainties. Hence, the impacts of changing raw material costs over a range of -80% to +60% of the base case scenario on the minimum biopropane selling price of biopropane have been evaluated, using a CO_2 selling prices of \$0/tonne and \$60/tonne [38] at 10% IRR. The results are presented in Figure 5 (a and b).

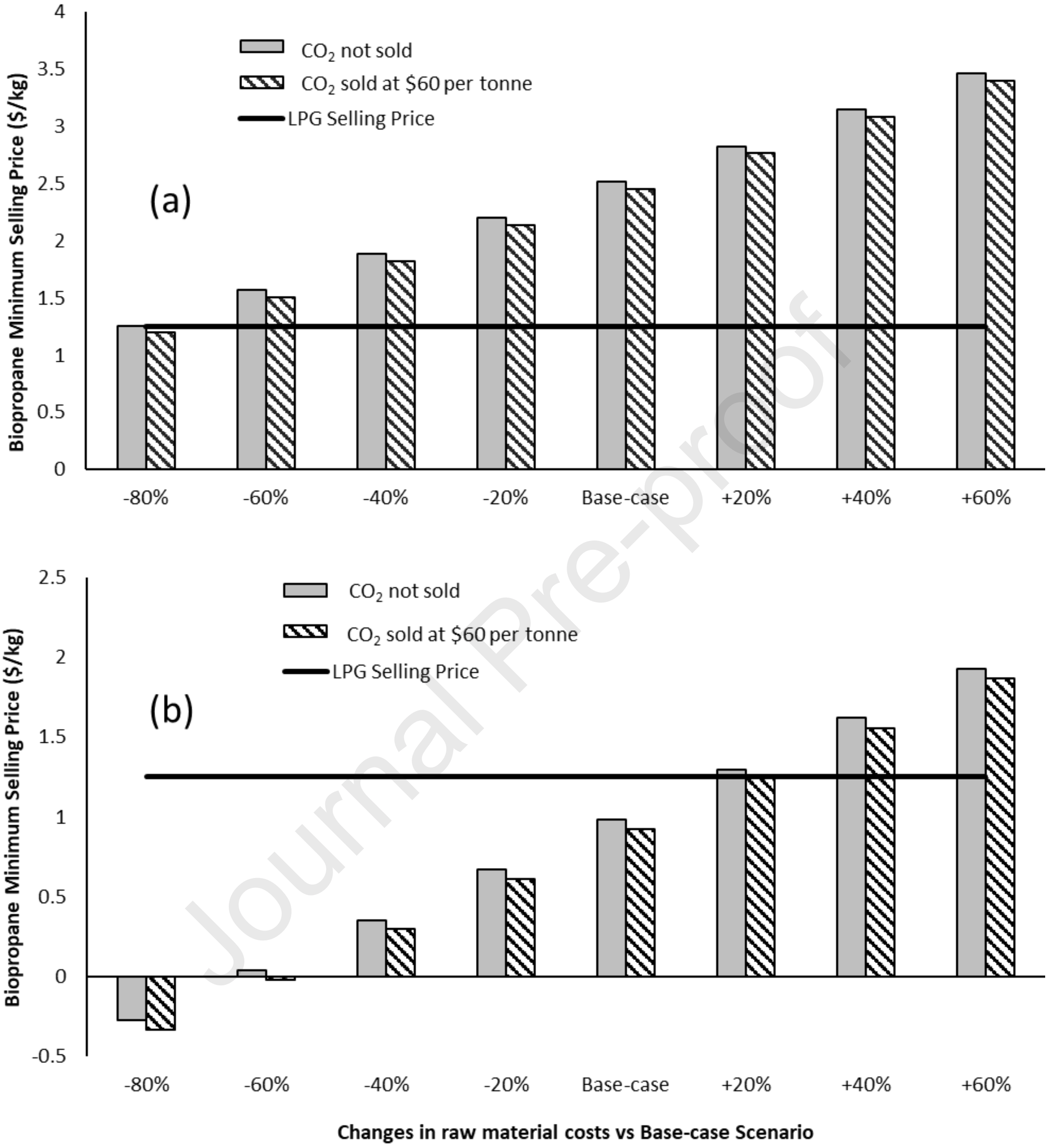

Figure 5: Effect of material costs on minimum selling price of biopropane at 20,000 tonnes per year production capacity (a) without RTCs and (b) with RTCs

Clearly, the trend in Figure 5a shows that the selling price has a positive correlation with material costs, with or without the sale of CO_2. For example, in the highly optimistic scenario of reducing the feedstock cost by 80%, the minimum biopropane selling price will be similar to the reference selling price of fossil LPG with values of $1.26/kg and $1.20/kg, depending respectively on whether CO_2 is sold or not and even without considering the impacts of incomes from RTCs. However, this is the only raw material cost scenario that can deliver selling price parity with fossil LPG, as any higher material costs considered led to biopropane minimum prices that are higher, when RTCs are not considered. Figure 5b clearly shows the significant impact that the incorporation of RTCs can have on the biopropane selling prices with respect to changing raw material costs. At 80% raw material cost reduction, the minimum selling prices of biopropane became negative, irrespective of whether CO_2 is sold (-$0.29/kg) or not (-$0.24/kg). The price still stayed negative at 60% raw material reduction if CO_2 is sold (-$0.02/kg). Indeed, Figure 5b shows that with incomes from RTCs, the minimum selling prices are much lower or similar to that of fossil LPG up to a 20% increase in the raw material cost in relation to the base case scenario. Therefore, with the incorporation of RTC incomes any reduction in raw material costs will deliver cheaper biopropane from the butyric acid route compared to the current price of fossil LPG. Indeed, with RTC incomes, the raw material costs will need to increase by over 20% of the base case value before the minimum selling price goes above $1.25/kg. Even then, at 60% increase in raw material costs, the minimum selling price is still under $2/kg, whether CO_2 is sold or not, which underlines the importance of incentivisation.

3.4.3 Effect of production capacity on minimum selling price of biopropane

Production capacity can have significant impact on production costs and therefore, selling prices of chemical products due to the concept of economy of scale, which is assumed in this study. In this present study, the effect of sequentially increasing production capacity from the base case 20,000 tonnes/year to 100,000 tonnes/year have been used for sensitivity analysis. The ISBL capital costs for each capacity have been scaled using the cost-to-capacity method, with an index factor, n = 0.6 (six-tenths rule) [29]. However, scaling up operating costs is more complicated; for example, the labour and supervision costs do not change much within the same order of magnitude of production capacity, whereas the other cost elements can change significantly. Hence, in this study each element of the operating costs has been analysed in detail, using the same methods used for the 20,000 tonnes/year base case. According to these analyses, doubling the production capacity from 20,000 tonnes/year to 40,000 tonnes/year reduced the production costs from \$2337.90/tonne to \$1652.32/tonne, therefore delivering biopropane at a cheaper cash cost than the HVO process (\$2312.51/tonne) [35]. Therefore, it is possible that this process, with relatively few processing steps can be favoured by economies of scale.

Using the total production costs obtained, the biopropane minimum selling prices at the different production capacities have been determined and presented in Figure 6. The analyses have been done without considering any incentives, and for two scenarios of selling CO_2 co-product at \$60/tonne and not selling it. The payback period and ROI averaged 9.06 years and 7.33 %, respectively when CO_2 is not sold. At a CO_2 selling price of \$60/tonne, the payback period reduced slightly to 9.03 years, with an average ROI of 7.2%.

Figure 6 shows that increasing the processing capacity will consistently deliver cheaper biopropane from this process, by taking advantages of the economies of scale. There is a much-pronounced drop when the production capacity is doubled with biopropane minimum selling prices reducing by about 29% for both scenarios. The drop in the minimum selling prices continued at high production capacities but a much slower rate.

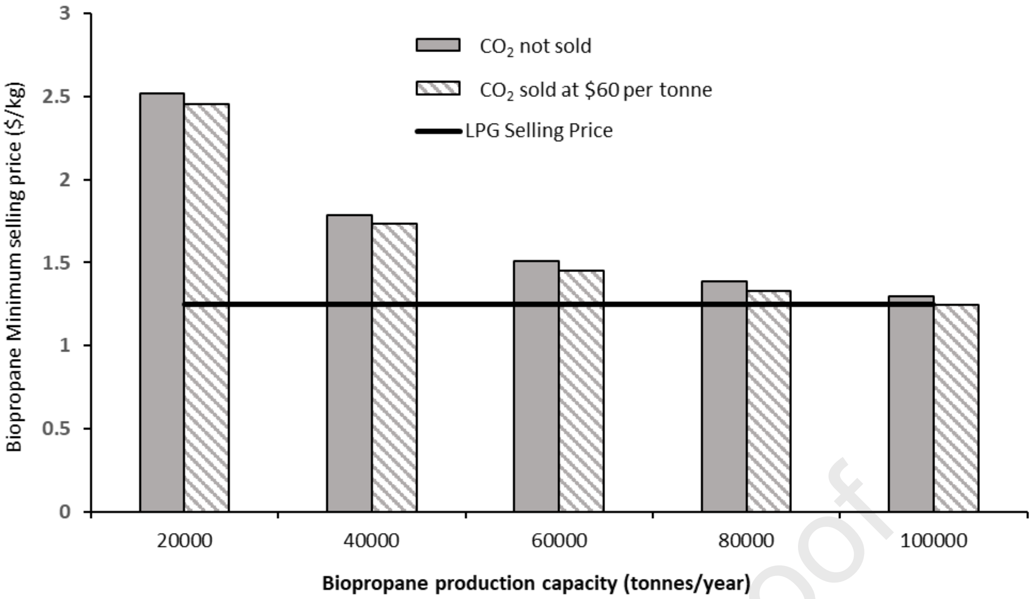

Figure 6: influence of biopropane production capacity on its minimum selling price

From this analysis, at a potential production capacity of 100,000 tonnes/year, this process could deliver biopropane at an incentive-free selling price of \$1.30/kg (without selling CO_2) and \$1.25/kg (with CO_2 sold at \$60/tonne). When incentives are included at the current rate of \$1.53/kg additional RTC incomes [4], this process has the potential to deliver much cheaper biopropane than the reference price of \$1.25/kg for fossil LPG, with negative selling prices at 60,000 tonne/year capacity and above. However, considerations must be given to availability of producing butyric acid feedstock, for example, at rate of 4.2 million tonnes to produce 100,000 tonnes of biopropane per year using this process. The physical and technical limitations of processing the amount of aqueous butyric acid feedstock required for such large capacity plants also need to be considered.

4.0 Conclusions

In this present study, a preliminary techno-economic study has been carried out on a hypothetical process to produce biopropane via the efficient catalytic decarboxylation of biomass-derived butyric acid. Using ASPEN Hysys, the process flowsheet has been developed using data from laboratory experiments, showing that the process is relatively simple. Process synthesis has also shown that the required pieces of equipment for the identified unit operations required for the development of the technology currently exists The ASPEN model, based on feedstock conversions and product yields, has been used to carry out economic analysis, using a hypothetic plant located in the UK and producing 20,000 tonnes/year of biopropane as the base case scenario. Results of the economic analysis indicate that the butyric acid process route can deliver biopropane at competitive selling prices to the reference fossil LPG product in the transport market, with the incorporation of RTC incomes. It can also be cost competitive to other biopropane production technologies.

There is still significant work ahead to prove that this novel technology can deliver biopropane at affordable costs to the end user, when produced at commercial scale. In addition, production of large quantities of butyric acid from biomass can become a bottleneck. It appears that significant investments in the production of butyric acid feedstock will be needed but the butyric acid technology has the advantage of using highly diluted feedstock, thereby eliminating the high costs of dewatering fermentation broths. The estimated production costs (capital and operating) have been based on a process model designed from laboratory experimental data. More research on the reaction kinetics, potential discovery of cheaper decarboxylation catalysts and the testing of the batch results in a laboratory-scale continuous reactor system will be important for the future development of a pilot-scale plant. Nonetheless, the results of this present study can contribute to the eventual commercialisation of this technology.

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Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: