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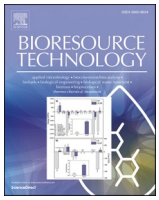


Techno-economic evaluation and life cycle assessment of a biorefinery using winery waste streams for the production of succinic acid and value-added co-products

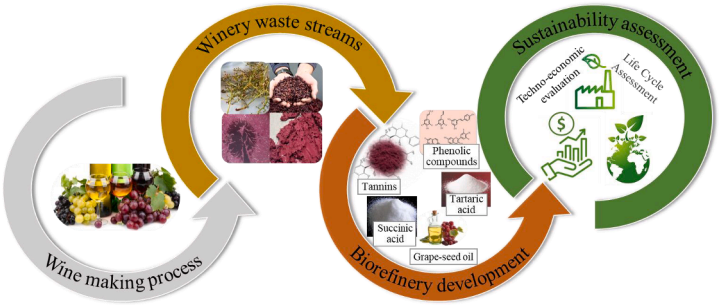
Sofia Maria Ioannidou [a], Katiana Filippi [a], Ioannis K. Kookos [b], Apostolis Koutinas [a], Dimitrios Ladakis [a,*]

[a] Department of Food Science and Human Nutrition, Agricultural University of Athens, Iera Odos 75, 118 55 Athens, Greece
[b] Department of Chemical Engineering, University of Patras, Rio, 26504 Patras, Greece

HIGHLIGHTS

- Biorefinery is developed using the three main winery waste streams.
- Techno-economic and LCA impact indicators were estimated.
- The co-product market prices affect biorefinery profitability.
- Cost-competitive succinic acid production could be achieved.

GRAPHICAL ABSTRACT



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ABSTRACT

This study presents techno-economic evaluation and life cycle assessment of a novel biorefinery using the three main waste streams generated by wineries for the production of bio-based succinic acid (SA), crude phenolic-rich extract, grape-seed oil, calcium tartrate and crude tannin-rich extract. Process design has been employed for the estimation of material and energy balances and the sizing of unit operations. The Minimum Selling Price of succinic acid production within a winery waste biorefinery ranges from \$1.23–2.76/kg_{SA} depending on the market price and the potential end-uses of the extracted fractions. The Global Warming Potential and the Abiotic Depletion Potential of winery waste valorisation through the proposed biorefinery are 1.47 kg CO_2-eq per kg dry waste and 25.2 MJ per kg dry waste, respectively. Biorefining of winery waste could lead to the development of a sustainable and novel bioeconomy business model with new market opportunities and efficient waste management.

* Corresponding author.
E-mail address: ladakisdimitris@gmail.com (D. Ladakis).

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1. Introduction

The global production of wine in 2018 was 29.2 million m^3 (OIV, 2019). The production of 0.7 L wine requires approximately 1 kg of grapes. Wineries generate significant quantities of waste streams. According to Galanakis (2017), the production of 1,000 m^3 of wine generates 82.5 t grape pomace (including skins and seeds) on a dry basis (db), 35.7 t grape stalks (db) and 85.7 t wine lees. Wine-making is a seasonal process and for this reason the residues produced should be processed in a short time. The disposal of such a large amount of waste causes environmental pollution problems because winery wastes are rich in phenolic compounds with a high organic load (Ahmad et al., 2020). Moreover, wineries may have to pay for waste disposal, while in many cases the cost is expended indirectly through the community (De Iseppi et al., 2020). In particular, wine lees disposal to the environment constitutes a major problem due to its high content in organic compounds at a low pH (De Iseppi et al., 2020). Biorefinery development is the only sustainable alternative for the valorisation of winery waste streams leading to the production of various bio-based products (Chowdhary et al., 2021; Sirohi et al., 2020).

Grape pomaces are usually discarded at a disposal cost, fermented to produce alcoholic beverages, employed in livestock feed production or used as fertiliser (Williams et al., 2019). Grape pomaces have been used for the production of various value-added products including enzymes, biogas, bioethanol, biopolymers, biochar and bio-active compounds among others (Chowdhary et al., 2021). Grape pomaces have been used for the extraction of bioactive compounds and grape-seed oil with applications in animal feed, pharmaceuticals, cosmetics and the food industry (Beres et al., 2017; Sirohi et al., 2020). The solids remaining after the extraction of bioactive compounds contain carbohydrates that could be used as carbon sources for the production of bio-based chemicals and polymers, such as polyhydroxyalkanoates (Martinez et al., 2016) and succinic acid (SA) (Filippi et al., 2021). Grape stalks have a low market value and are either discarded or used as fertilizers, while they could be used as carbon sources in fermentation processes (Filippi et al., 2021). The wine lees produced in the clarification process could be used for the extraction of bioactive compounds, ethanol, calcium tartrate and yeast cells that could be converted into a nitrogen-rich fermentation supplement (Dimou et al., 2016).

Literature-cited studies on biorefinery development have mainly used individual winery waste streams (Dimou et al., 2016; Ahmad et al., 2020). Filippi et al. (2022) proposed the utilisation of all major winery waste streams for the production of multiple end-products. In this way, conventional wineries could be restructured into sustainable biorefineries. The integration of bio-based chemical production in such biorefineries is the only way to achieve their sustainable production that cannot be achieved by conventional bioprocesses. For instance, bio-based succinic acid production in industrial facilities is currently carried out by Myriant, Reverdia, Succinity and LCY Biosciences. The production of bio-based succinic acid is not cost-competitive as compared to petro-based succinic acid due to high capital investment requirements, technology issues, economies of scale requirements, adequate supply of raw materials and demanding R&D to deliver a sustainable product (MarketsAndMarkets, 2021). The main carbon sources used for bio-based succinic acid production are glucose syrup and glycerol using engineered bacterial or yeast strains (e.g. *Actinobacillus succinogenes*, *Basfia succiniciproducens*, *Escherichia coli*). The integration of bio-based succinic acid production in novel biorefineries using crude renewable resources could lead to process sustainability as compared to petro-based succinic acid (Babaei et al., 2019; Li et al., 2019; Stylianou et al., 2021). Filippi et al. (2022) showed that grape pomace, grape stalks and wine lees could be employed in a novel biorefinery concept for the production of both high value – low volume products (e.g. polyphenols) and low value – high volume products (e.g. succinic acid).

This study focuses on the techno-economic evaluation (TEA) and life cycle assessment (LCA) of the novel biorefinery presented by Filippi et al. (2022) using winery waste for the production of bio-based succinic acid and value-added co-products, namely crude phenolic-rich extract (CPE), grape-seed oil (GO), calcium tartrate (CaT) and crude tannin-rich extract (CTE). Previous studies have carried out techno-economic analysis to evaluate the profitability potential of either single product generation from winery waste or the valorisation of a single winery waste stream. Dimou et al. (2016) carried out a techno-economic evaluation of wine lees valorisation to produce ethanol, calcium tartrate, antioxidants and yeast cells as animal feed. Jin et al. (2021) presented a techno-economic evaluation for the production of grape-seed oil, polyphenols and biochar from grape pomace. Todd and Baroutian (2017) presented a techno-economic evaluation for the extraction of bioactive compounds from grape pomace utilising different extraction techniques. Duba and Fiori (2019) evaluated the economic feasibility of grape-seed oil extraction. This study assesses the holistic valorisation of all major winery waste streams and the potential reduction in succinic acid production cost through integrated biorefinery development.

2. Materials and methods

2.1. Biorefinery description and design

The proposed biorefinery involves three different winery waste streams, namely grape pomaces, which include skins and seeds, grape stalks and wine lees generated after the winemaking process. All design parameters (e.g. processing conditions, extraction yields, pretreatment and hydrolysis yields, fermentation efficiency, material balances) for the proposed biorefinery have been taken from Filippi et al. (2022). All process flowsheets described below have been developed using the experimental results presented by Filippi et al. (2022). Succinic acid is produced via fermentation using the carbohydrate content of waste streams after the extraction of value-added fractions. The process design software UniSim (Honeywell) has been used to carry out all simulations.

In order to determine the annual waste utilization and co-product generation of the biorefinery, the annual production of succinic acid was set at around 30,250 t/y. This value is a common annual production quantity of a platform chemical at which economies of scale can be achieved (Bonatsos et al., 2020; Stylianou et al., 2021). Given the succinic acid production capacity mentioned above, the carbohydrate content of winery wastes and the conversion yields and fermentation efficiency reported by Filippi et al. (2022), the required quantity of grapes (2.48 million t/y) and the resulting wine production capacity (1.73 million t/y) were estimated. Based on these quantities, the generated winery wastes were estimated as 805,536 t/y containing 77% grape pomace, 12% grape stalks and 11% wine lees. Grape pomace and stalks have moisture contents of 75% and 50%, respectively, while the solid content of wine lees is 20.8% (w/w) (Galanakis, 2017; Ioannidou et al., 2020). Fig. 1 presents the material balances of the proposed biorefinery concept using the experimental results presented by Filippi et al. (2022) and the quantities of individual waste streams and succinic acid production presented above.

Due to seasonal production of wine, it is assumed that the wastes are stored so that they can be used throughout the year to ensure the continuous operation of the plant. After extraction of free sugars, the grape pomace is dried prior to storage until further processing.

2.1.1. Grape pomace processing (Area 100)

As illustrated in Fig. 2A, soluble sugars contained in grape pomace (skin and seeds) are initially extracted with water at 40°C for 2 h (V-101) under continuous stirring (A-101). The solid residue is separated from the slurry via centrifugation (CF-101) and the obtained liquid stream is fed to a mechanical vapour recompression (MVR) – forced circulation evaporator system (EV-103, C-103, E-103) to concentrate the free sugar fraction to 500 g/L. The concentration of the free sugars stream facilitates storage for longer periods until the free sugars are used as carbon

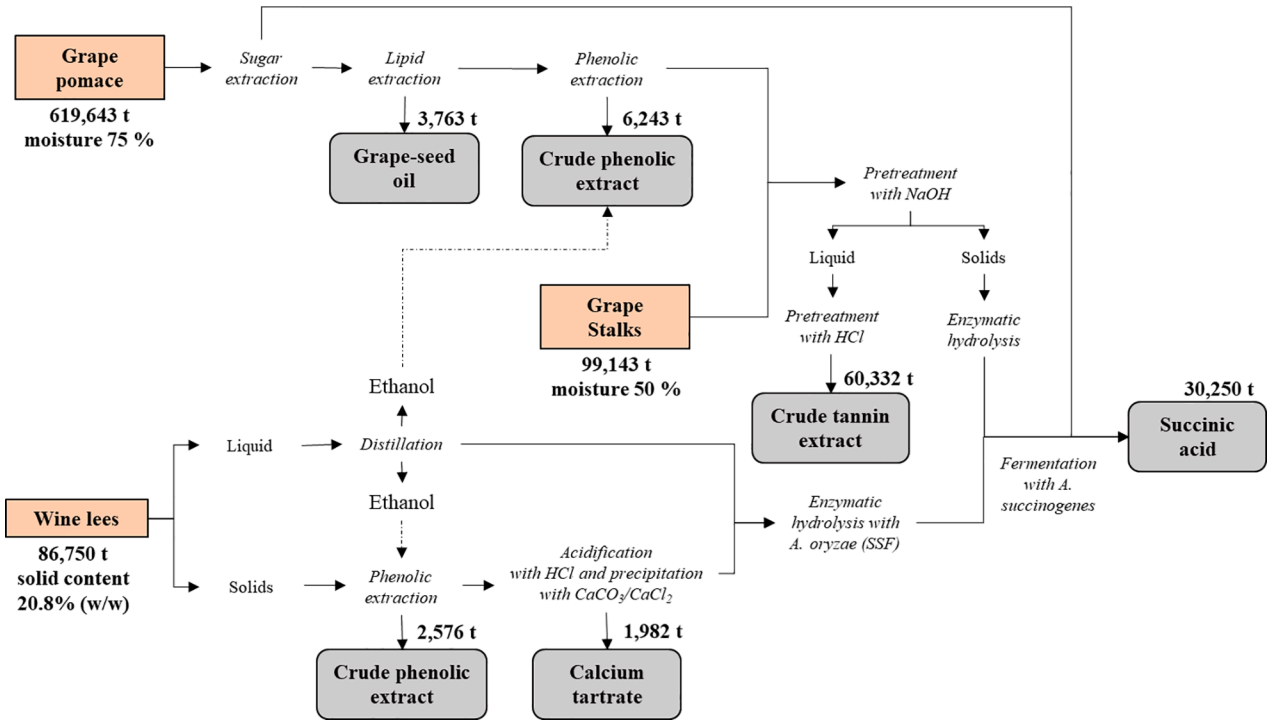

Fig. 1. Mass balances in the proposed biorefinery using winery waste streams for the extraction of value-added fractions and the production of bio-based succinic acid via fermentation.

source for succinic acid production in Area 400. The solid stream after centrifugation (stream 102) is dried (DR-101) and stored to facilitate storage for longer periods until further processing.

The dried solids (stream 103) are fed into the vessel V-102 for GO extraction with ethyl-lactate under continuous agitation (A-102) for 2 h at ambient temperature. The suspension is centrifuged (CF-102) to separate the solid from the liquid fraction. The GO is isolated by evaporation (EV-101) under vacuum at 70°C. The recovered ethyl-lactate is recycled in the GO extraction vessel (V-102), while 5% ethyl-lactate is added to replace the losses of the solvent during processing. The phenolic compounds contained in the remaining solid fraction (stream 105) are extracted with 70% (v/v) aqueous ethanol for 20 min at 1:10 solid-to-liquid ratio (V-103). The ethanol used for CPE extraction has been extracted from wine lees in Area 300 (Fig. 3A). This is an important sustainability aspect in the proposed biorefinery as no commercial ethanol supply is required for CPE extraction. The centrifugal separator CF-103 is employed to separate the liquid stream, which is concentrated in the evaporator EV-102 under vacuum at 40°C for the recovery of the CPE. The ethanol solution is recycled in the extraction vessel V-103, while the remaining solids (stream 107) are directed to Area 200 for further treatment (Fig. 2B).

2.1.2. Grape stalks processing (Area 200)

Fig. 2B presents the grape stalks treatment process. Stream 107 from grape pomace processing (Area 100) is mixed with grape stalks in V-201. The mixture is subjected to dilute aqueous (1.19%, w/v) sodium hydroxide pretreatment at 1:10 solid-to-liquid ratio and 30 min residence time. The mixed effluent is centrifuged (CF-201) and the liquid stream 202 is treated with 3 N HCl for 10 min in V-202 for CTE precipitation. The precipitated tannin-rich crude fraction is separated via centrifugation (CF-202) and dried (DR-201).

The solid residue (stream 205) obtained after centrifugation (CF-201) is fed into a mixing tank (V-203) and the pH is adjusted with dilute HCl. The slurry is fed into a vessel (V-204) together with water to achieve a solid concentration of 10% (w/v) and enzymes to hydrolyse the cellulose and hemicellulose fractions. The enzymatic hydrolysis is conducted for 48 h using the experimental results reported by Filippi et al. (2022). The sugar-rich hydrolysate is separated via centrifugation (CF-203) and subsequently used in the fermentation stage (Area 400).

2.1.3. Wine lees processing (Area 300)

Area 300 presents the process flow diagram of wine lees fractionation (Fig. 3A). A centrifugal separator (CF-301) separates the solids from the liquid fraction. A distillation column (T-301) is then employed for ethanol recovery from stream 301. The recovered ethanol is used for CPE extraction in the biorefinery to eliminate the use of commercial ethanol. The phenolic compounds contained in solid lees are extracted with 50% (v/v) aqueous ethanol in a mixing tank (V-301) for 1 h. The slurry is directed towards a centrifugation step (CF-302). The liquid stream is fed into the evaporator EV-301 to separate the ethanol/water mixture from the CPE. After centrifugation, the solid stream 305 is transferred to a mixing tank (V-302) and suspended in water and HCl to separate the tartaric acid. After 10 min of continuous stirring, the tartaric acid-rich solution is separated from the solids by centrifugation (CF-303). Stream 311 is mixed with $CaCO_3$ and $CaCl_2$ to transform the soluble tartaric acid into the insoluble calcium tartrate according to the process presented by Dimou et al. (2016). Calcium tartrate is separated from the liquid in CF-306 and the solid stream 313, containing 50% solids, is dried (DR-301) to obtain the final product.

Sunflower meal (SFM) is used as a solid substrate in the solid-state fermentation (TF-301) for the production of crude enzymes (mainly protease) using the fungal strain *Aspergillus oryzae,* as previously described by Kachrimanidou et al. (2021). The crude enzyme consortia are produced (TF-301) at a moisture content of 65% (w/w, on a wet basis). After 48 h, the whole solid state fermentation solids that contain the crude enzymes are mixed (V-304) with the aqueous liquid stream 308 obtained after ethanol distillation. The mixture is then centrifuged (CF-304) and the liquid stream 309, containing the crude enzymes, is fed in V-305 along with the wine lees solid stream 307. The enzymatic hydrolysis is carried out for 48 h at 40°C. The pH is adjusted with NaOH. After hydrolysis completion, the liquid stream, rich in free amino nitrogen (FAN), is separated via centrifugation (CF-305). The FAN-rich

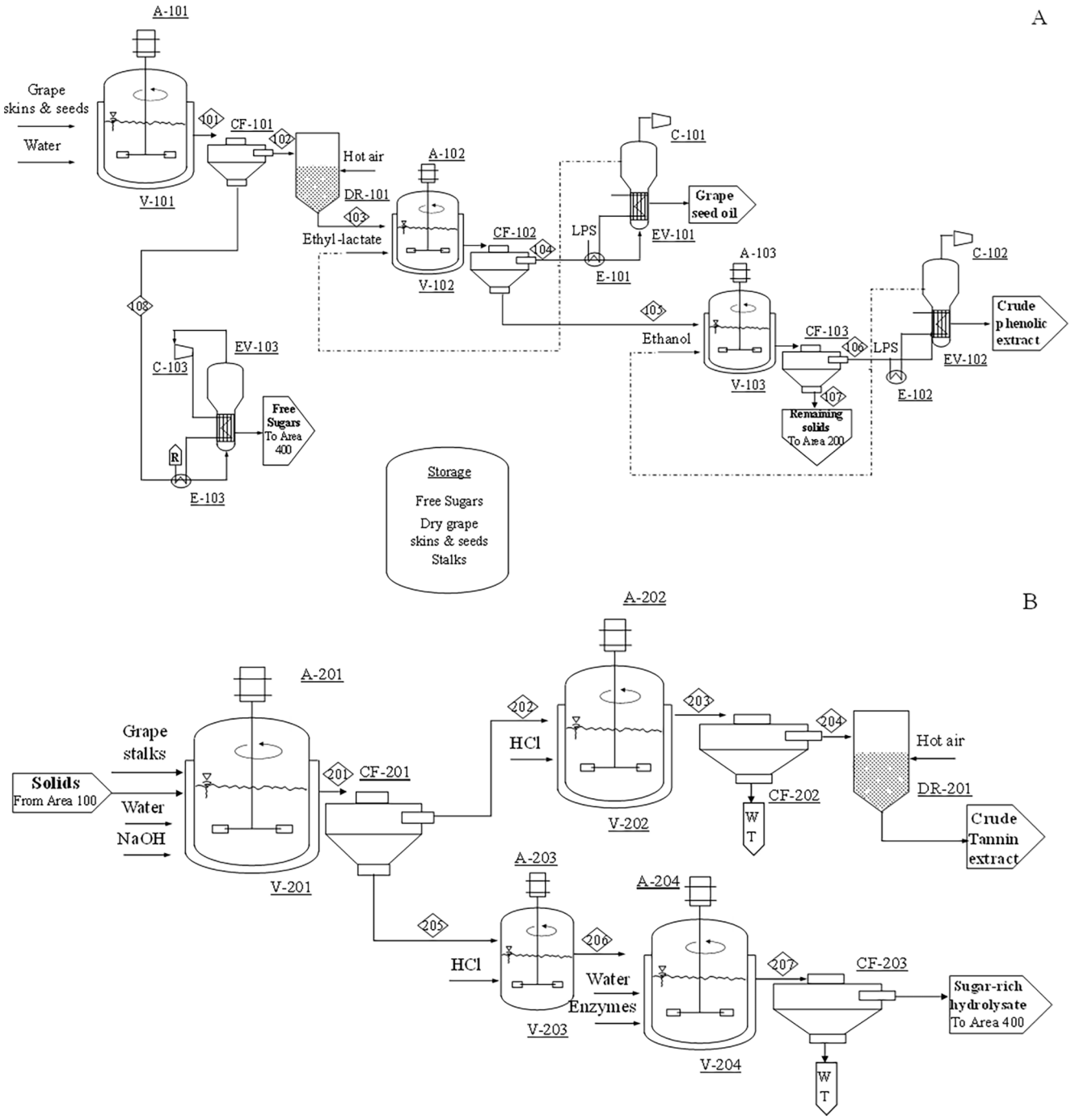

Fig. 2. Process flow diagram of grape pomace (Area 100) (A) and grape stalks (Area 200) (B) processing.

hydrolysate is used as fermentation nutrient supplement in Area 400.

2.1.4. Succinic acid production (Area 400)

Fig. 3B presents the fermentative production of succinic acid as well as its downstream separation and purification (DSP) stages. The concentrated free sugars extracted from grape pomace (Area 100), the sugar-rich hydrolysate (Area 200) and the FAN-rich hydrolysate produced via enzymatic hydrolysis of wine lees (Area 300) are mixed (V-401) and sterilized in continuous operation mode using three heat exchangers (E-401, E-402, E-403). The sterilized stream is fed into the bioreactor F-403. Succinic acid production is carried out with the bacterial strain *A. succinogenes* at 37°C under continuous sparging of CO_2. The inoculation bioreactor train (F-401, F-402) is used for inoculum preparation. After 47 h, 37.2 g/L succinic acid are produced with 0.64 g/g sugar to succinic acid conversion yield and 0.79 g/(L•h) productivity. Succinic acid crystals are subsequently purified using the DSP described by Alexandri et al. (2019). The fermentation broth is centrifuged (CF-401) to remove the bacterial biomass. Stream 403 is fed to activated carbon columns (V-402) to achieve decolorisation and to remove impurities. The decolorized effluent is fed into cationic resin columns (V-403) to transform organic acid salts into their corresponding organic acids. The acidified liquid stream is then mixed with the liquid stream that is recycled from the crystallizers (V-404) and the resulting stream is concentrated using the MVR - forced circulation evaporator system (EV-401). Stream 404 is subsequently treated via crystallization in continuous crystallizers (CR-401, CR-402) at 4°C. The remaining liquid after

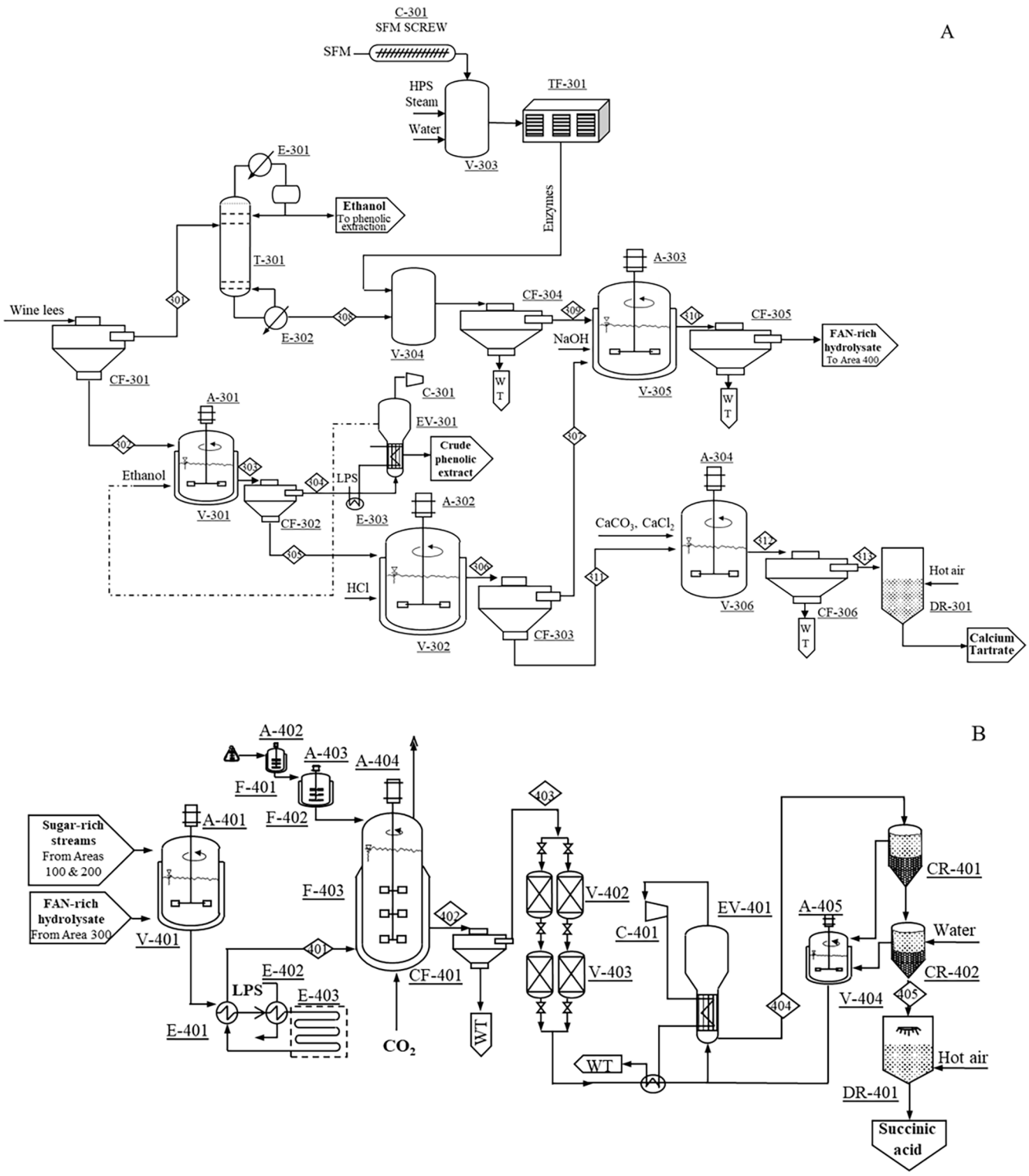

Fig. 3. Process flow diagram of wine lees processing (Area 300) (A) and bio-based succinic acid production (Area 400) (B).

crystallization is recycled to the evaporation stage. Dried succinic acid crystals are produced using a spray dryer (DR-401). The succinic acid crystal purity achieved is higher than 99.5%, while the overall succinic acid recovery yield in the DSP is ca. 95% (w/w).

2.2. Techno-economic evaluation

The mass and energy balances were estimated using the experimental results presented by Filippi et al. (2022). Sizing of unit operations was performed based on standard engineering procedures described in the literature (Peters et al., 2003; Ulrich and Vasudevan, 2004; Turton et al., 2018). The preliminary techno-economic methodology followed with accuracy up to ± 30% has been described by Ioannidou et al. (2021). The industrial plant was assumed to operate 7,920 h/y. Fixed Capital Investment (FCI) was estimated by multiplying the sum of the free-on-board purchased equipment costs (Ceq.fob) with

a Lang factor of 5 (Dheskali et al, 2020). The Lang factor ranges from 3 to 6 when the Ceq.fob is used for the estimation of the FCI (Peters et al., 2003; Turton et al., 2018). A Lang factor of 5 has been used in this study due to the construction of a new industrial plant using a high-risk new technology and the utilisation of expensive construction material in the process equipment.

The cost of manufacture (COM) was estimated using the equation proposed by Turton et al. (2018).

$$COM = 0.18 \times FCI + 2.73 \times C_{OL} + 1.23 \times (C_{UT} + C_{RM}) \quad \text{Eq.1}$$

where C_{OL} represents the operating labour cost, C_{UT} includes utilities expenses and C_{RM} stands for the raw material expenses.

The coefficients used in equation 1 have been estimated by Turton et al. (2018) in order to include the contribution of all secondary product manufacturing cost categories (e.g. maintenance, marketing, R&D) that can be associated to the main cost categories used in equation 1. The unitary cost of utilities, supplied by off-sites, are $9.45/t low pressure steam (LPS), $0.0674/kWh electricity and $0.0157/t cooling water (Turton et al., 2018). The C_{UT} is estimated by multiplying the unitary cost of each utility with the utility requirements presented in Table 1. The methodology reported by Ulrich and Vasudevan (2004) has been employed to estimate the C_{OL} considering the total number of workers required in the industrial plant based on the annual plant operation (Table 2), the working time of each worker and the average labour cost. The C_{RM} is estimated by multiplying the unitary raw material costs with the raw material requirements presented in Table 3.

In EU, the solid waste disposal cost could be as low as $35/t (Hogg, 2002). The transportation cost of raw grape pomace is ca. $32/t (Jin et al., 2021). Taking into consideration the lowest solid waste disposal cost ($35/t) that should be paid by the winery producing the winery wastes and the transportation cost ($32/t) that should be paid by the biorefinery to transport the winery waste to the facility, a zero cost has been considered for the winery waste to be used as feedstock in the biorefinery.

Discounted cash flow (DCF) analysis has been carried out using the parameters reported by Humbird et al. (2011), namely 10% interest rate, 30 years plant lifetime, 100% equity financing, 7 years for depreciation based on the Modified Accelerated Cost Recovery System (MARS), 3 years plant construction period and working capital estimation as 5% of the FCI. The TEA indicators used in this study are the Net Present Value (NPV), the Discounted Payback Period (DPP) and the Minimum Selling Price (MSP), while emphasis has been given on the feedstock availability based on the wine production in the major EU wine producing countries.

Table 1
Life cycle inventory for the proposed biorefinery using winery waste streams.

Inputs		Outputs	
Raw material / Utility	**Value**	**Product**	**Value**
Pomace (t)	619,643	Succinic acid (t)	30,250
Stalks (t)	99,143	Grape-seed oil (t)	3,763
Lees (t)	86,750	Crude phenolic extract (t)	8,819
Ethyl-lactate (t)	5,553	Calcium tartrate (t)	1,982
NaOH (t)	22,189	Crude tannin extract (t)	60,332
HCl (t)	203,702		
$MgCO_3$ (t)	4,066		
CO_2 (t)	16,919		
Other nutrients (t)	2,536		
$CaCO_3$ (t)	1,059		
$CaCl_2$ (t)	1,059		
Enzymes (t)	681		
SFM (t)	107		
Electricity (kWh)	292,823,588		
Steam (t)	599,751		
Water (t)	1,931,283		

2.3. Life cycle assessment

Environmental life cycle assessment was carried out based on the LCA principles, a standardized methodology for the environmental assessment according to the ISO 14040 and 14044 standards (ISO, 2006). This framework is divided into the goal and scope definition, the inventory analysis, the impact assessment and the interpretation of results. The scope of this study along with the functional unit and system boundaries are defined in the first stage, while in inventory analysis, the input and output data of all areas are determined. The impact assessment phase reports the selected assessment methodology and the impact categories that will be analysed. Finally, the LCA results are discussed and compared to relevant published studies in the interpretation phase. In this study, life cycle assessment was carried out using the LCA software GaBi for the estimation of environmental indicators.

2.3.1. Goal and scope

The aim of this study is to assess the environmental performance of a biorefinery using winery wastes. A “cradle-to-gate” LCA approach has been employed for the analysis, considering as functional unit 1 kg of dry waste stream after the production of 2.15 kg wine. The composition of 1 kg wet waste is 77% grape skins and seeds (75% moisture content), 12% grape stalks (50% moisture content) and 11% wine lees (20.8% solid content). The system boundaries for the LCA includes the treatment of grape pomace (skins and seeds) for the production of GO and CPE as well as the extraction of the free sugars, the treatment of stalks for the extraction of CTE and the production of a sugar-rich hydrolysate, the wine lees treatment for the production of CPE, CaT and a FAN-rich hydrolysate, and finally succinic acid production and purification.

2.3.2. Life cycle inventory (LCI)

The life cycle inventory that includes the mass and energy inputs and outputs of the whole biorefinery is presented in Table 1. The construction of the inventory was based on the developed process flow diagrams. The presented quantities are related to the treatment of 805,536 t of wet winery waste. After the analysis, the results have been expressed to the selected functional unit. Electricity generation from grid and steam generation using natural gas have been considered in this study. The environmental impact of the winery wastes was taken from Fusi et al. (2014) where a “cradle-to-grave” LCA was presented to estimate the environmental performance of 750 mL Sardinian white wine production. Fusi et al. (2014) implemented economic allocation to distribute the environmental impacts among the main product (wine) and the waste streams. The environmental impact has been estimated as 8.21×10^{-4} kg CO_2-eq per kg grape pomace and wine lees, while no impact was attributed to grape stalks. It should be mentioned that carbon sequestration via grape cultivation and the release of CO_2 during wine fermentation have not been taken into consideration in the LCA conducted in this study.

2.3.3. Life cycle impact assessment

In this study, the LCA was carried out using the CML 2001 (Jan. 2016) methodology (Guinée et al., 2002; Ioannidou et al., 2020). The final results are expressed using the quantitative indicators Global Warming Potential (GWP) and Abiotic Depletion Potential (ADP fossil) (Ioannidou et al., 2020).

3. Results and discussion

3.1. Techno-economic evaluation

3.1.1. Fixed Capital Investment estimation

Table 2 presents the Ceq.fob for all process equipment employed in grape pomace processing containing skins and seeds (Area 100), grape stalks processing (Area 200), wine lees processing (Area 300) and succinic acid production (Area 400). In Area 100, the main purchase

Table 2
Purchase equipment cost, FCI, cost of operating labor (C_{OL}) and cost of utilities (C_{UT}) for the proposed biorefinery.

	Unit	Description	Unit number	$CEPCI_{t0}$	Characteristic size (X_t)	FOB Cost (C_p@2018, M$)	Electricity (kWh/y)	Steam (t/y)	Process water (t/y)
Area 100	V-101	Mixing tank[c]	1	521.9	V = 2,133.7 m^3	0.550			
	A-101	Agitator[a]	1	521.9	P = 254.34 hp	0.209	1,502,164.5		
	CF-101	Centrifugal separator[b]	3	444.2	Q = 71.12 m^3/h	2.123	736,560.0		
	EV-103	Evaporator[c]	6	521.9	A = 855.61 m^2	2.501	15,349,000.6		
	C-103	Compressor[a]	1	521.9	P = 1,744.2 kW	0.448			
	E-103	Heat exchanger[a]	9	444.2	A = 906.06 m^2	3.171			
	DR-101	Dryer[c]	1	525.4	A = 7.85 m^2	0.367	7,189,418.8	2,761.1	
	V-102	Mixing tank[c]	1	521.9	V = 371.27 m^3	0.313			
	A-102	Agitator[a]	1	521.9	P = 442.56 hp	0.362	2,613,766.2		
	CF-102	Centrifugal separator[b]	1	444.2	Q = 49.50 m^3/h	0.546	736,560.0		
	EV-101	Evaporator[c]	1	521.9	A = 57 m^2	0.397			
	C-101	Compressor[a]	1	521.9	P = 1,191.8 kW	0.373	10,488,178.7		
	E-101	Heat exchanger[a]	1	444.2	A = 47.65 m^2	0.039		70,602.5	
	V-103	Mixing tank[c]	1	521.9	V = 88.44 m^3	0.205			
	A-103	Agitator[a]	1	521.9	P = 105.42 hp	0.093	622,648.7		
	CF-103	Centrifugal separator[b]	3	444.2	Q = 70.82 m^3/h	2.116	736,560.0		
	EV-102	Evaporator[c]	1	521.9	A = 544.93 m^2	0.426			
	C-102	Compressor[a]	6	521.9	P = 2,600.0 kW	3.254	137,281,842.2		
	E-102	Heat exchanger[a]	1	444.2	A = 33.63 m^2	0.034		58,712.7	
	Unitary utility cost						$0.0674/kWh	$9.45/t	$0.0154/t
	Workers A100 = 7								
	Total Ceq.fob (M$)					**17.531**			
	FCI A100 (M$)			**5 × Total Ceq.fob A100 =**		**87.653**		**C_{UT} A100 (M$/y) =**	**13.680**
Area 200	V-201	Mixing tank[a]	1	521.9	V = 18.93 m^3	0.026		175,300.6	
	A-201	Agitator[a]	1	521.9	P = 22.57 hp	0.026	133,293.5		
	CF-201	Centrifugal separator[b]	1	444.2	Q = 30.29 m^3/h	0.403	237,600.0		
	V-202	Mixing tank[a]	1	521.9	V = 49.73 m^3	0.026			
	A-202	Agitator[a]	1	521.9	P = 59.27 hp	0.056	350,082.2		
	CF-202	Centrifugal separator[b]	3	444.2	Q = 79.55 m^3/h	1.702	736,560.0		
	DR-201	Dryer[c]	1	525.4	A = 5.00 m^2	0.220	1,877,409.4	721.0	
	V-203	Mixing tank[a]	1	521.9	V = 4.89 m^3	0.026			
	A-203	Agitator[a]	1	521.9	P = 5.84 hp	0.013	34,477.5		
	V-204	Mixing tank[a]	1	521.9	V = 163.18 m^3	0.042		51,514.2	
	A-204	Agitator[a]	1	521.9	P = 194.51 hp	0.162	1,148,813.1		
	CF-203	Centrifugal separator[b]	2	444.2	Q = 65.27 m^3/h	1.328	736,560.0		
	Unitary utility cost						$0.0674/kWh	$9.45/t	$0.0154/t
	Workers A200 = 3								
	Total Ceq.fob (M$)					**4.030**			
	FCI A200 (M$)			**5 × Total Ceq.fob A200 =**		**20.150**		**C_{UT} A200 (M$/y) =**	**2.866**
Area 300	CF-301	Centrifugal separator[b]	1	444.2	Q = 12.61 m^3/h	0.271	237,600.0		
	T-301	Distillation column[c]	1	240.0	N = 22	0.167			
	E-301	Heat exchanger[a]	1	444.2	A = 25.91 m^2	0.027			1,098,950.9
	E-302	Heat exchanger[a]	1	444.2	A = 38.42 m^2	0.031		31,800.2	
	V-301	Mixing tank[a]	1	521.9	V = 34.33 m^3	0.026			
	A-301	Agitator[a]	1	521.9	P = 40.93 hp	0.042	241,711.8		
	CF-302	Centrifugal separator[b]	1	444.2	Q = 27.47 m^3/h	0.382	237,600.0		
	EV-301	Evaporator[c]	1	521.9	A = 65.45 m^2	0.402			
	C-301	Compressor[a]	1	521.9	P = 1,873.6 kW	0.464	16,488,554.4		
	E-303	Heat exchanger[a]	1	444.2	A = 73.57 m^2	0.049		128,433.2	
	V-302	Mixing tank[a]	1	521.9	V = 3.10 m^3	0.202			
	A-302	Agitator[a]	1	521.9	P = 4.16 hp	0.013	5,697.7		
	CF-303	Centrifugal separator[b]	1	444.2	Q = 4.05 m^3/h	0.207	118,800.0		
	V-303	Mixing tank[a]	1	521.9	V = 14.07 m^3	0.202			
	TF-301	Tray SS bioreactors[b]	1	390.4	A = 112.59 m^2	0.105			

(*continued on next page*)

Table 2 (*continued*)

	Unit	Description	Unit number	$CEPCI_{t0}$	Characteristic size (X_t)	FOB Cost (C_p@2018, M$)	Electricity (kWh/y)	Steam (t/y)	Process water (t/y)
	CF-304	Centrifugal separator[b]	1	444.2	Q = 7.22 m^3/h	0.230	118,800.0		
	V-305	Mixing tank[c]	1	521.9	V = 495.89 m^3	0.128		1,969.9	
	A-303	Agitator[a]	1	521.9	P = 591.09 hp	0.488	3,491,056.9		
	CF-305	Centrifugal separator[b]	1	444.2	Q = 8.26 m^3/h	0.238	118,800.0		
	V-306	Mixing tank[a]	1	521.9	V = 2.05 m^3	0.197			
	A-304	Agitator[a]	1	521.9	P = 2.44 hp	0.013	14,390.0		
	CF-306	Centrifugal separator[b]	1	444.2	Q = 0.82 m^3/h	0.183	118,800.0		
	DR-301	Dryer[c]	1	525.4	A = 1.12 m^2	0.170	246,705.6	94.7	
	Unitary utility cost						$0.0674/kWh	$9.45/t	$0.0154/t
	Workers A300 = 5								
	Total Ceq.fob (M$)					**4.199**			
	FCI A300 (M$)			**5 × Total Ceq.fob A300 =**		**20.997**	**C_{UT} A300 (M$/y) =**		**4.084**
Area 400	E-401	Heat exchanger[a]	2	444.2	A = 823.77 m^2	0.639			
	E-402	Heat exchanger[a]	1	444.2	A = 86.28 m^2	0.054			
	E-403	Holding tube[a]	1	500	l = 0.12 *m*	0.146		16,341.8	
	F-403	Bioreactor[a]	11	521.9	V = 645.79 m^3	6.899			
	A-404	Agitator[a]	11	521.9	P = 769.78 hp	7.047			
	F-401/402	Seed bioreactor[a]	1	521.9	V = 64.58 m^3	0.187			5,603,523.4
	A-401/402	Seed agitator[a]	1	521.9	P = 76.98 hp	0.070	42,466,129.1		
	CF-401	Centrifugal separator[b]	2	444.2	Q = 52.92 m^3/h	1.143	868,604.2		
	V-402/V-403	I.E. resins[b]	2	521.9	V = 53.18 m^3	0.671			
	EV-401	Evaporator[c]	1	521.9	A = 1,195.9 m^2	1.639	24,389,732.2	10,851.9	
	CR-401	Crystalizer[c]	1	525.4	M = 24,519.90 kg/h	1.011	5,861,653.8		
	CR-402	Crystalizer[c]	1	525.4	M = 9,090.38 kg/h	0.616	246,076.1		
	DR-401	Dryer[c]	1	525.4	M = 424.39 kg/h	5.119	18,044,328.1		
	Unitary utility cost						$0.0674/kWh	$9.45/t	$0.0154/t
	Workers A400 = 19								
	Total Ceq.fob (M$)					**25.163**			
	FCI A400 (M$)			**5 × Total Ceq.fob A400 =**		**125.814**	**C_{UT} A400 (M$/y) =**		**6.591**
Total	**Total FCI A100-400 (M$)**					**254.66**			
	Total C_{OL} A100-400 (M$)					**6.36**			
							Total C_{UT} A100-400 (M$)		**27.221**

[a] Dheskali et al., 2017,
[b] Peters et al., 2003,
[c] Turton et al., 2018.

Table 3
Raw materials cost (C_{RM}) for the proposed biorefinery using winery waste.

Material	Amount (t/y)	Unitary cost ($/t)	Total cost (M$/y)
Ethyl-lactate [a]	5,552.6	1,110	6.163
NaOH [b]	22,188.7	400	8.875
HCl [b]	203,702.3	61	12.426
$MgCO_3$ [b]	4,065.9	1,000	4.066
CO_2 [b]	16,919.5	150	2.538
Other nutrients [b]	2,536.5		1.633
$CaCO_3$ (t) [c]	1,059.0	150	0.159
$CaCl_2$ (t) [c]	1,059.0	150	0.159
Enzymes [d]	681.5	4,210	2.869
SFM [e]	107.0	250	0.027
Process water [f]	1,931,283.1	0.435	0.839
Total C_{RM} (M$)			39.75

[a] Alibaba.com (2021),
[b] ICIS - Independent Commodity Intelligence Services (2021),
[c] Dimou et al. (2016)
[d] Humbird et al. (2011),
[e] Kachrimanidou et al. (2021),
[f] Turton et al. (2018)

equipment costs are attributed to the evaporator systems employed for sugar concentration, grape-seed oil extraction and CPE extraction. In Area 200, the centrifugal separators CF-202 and CF-203 contribute the highest percentage of purchase equipment costs, reaching 42% and 33%, respectively. In Area 300, the evaporator system for CPE extraction from the solid fraction of wine lees contributes ca. 22% of the total purchase equipment cost in this section. In Area 400, where succinic acid crystals are produced, the bioreactors and agitators used contribute the highest percentage in the purchase equipment cost (ca. 56%), while the second highest purchase cost is attributed to the dryer DR-401 (20%). The overall FCI for the whole biorefinery is M$254.7. The FCI of the succinic acid production section (Area 400) contributes around 50% of the total FCI of the whole biorefinery.

3.1.2. Estimation of Cost of Manufacture

The COM (M$145.6) of the whole biorefinery presented in Figs. 2 and 3 has been estimated considering 30,250 t annual succinic acid production using 805,536 t/y winery waste containing 77% grape pomace, 12% grape stalks and 11% wine lees on wet basis. The COM has been estimated using the C_{UT} (M$27.2), the C_{OL} (M$6.4) and the FCI (M $254.7) presented in Table 2 for Areas 100–400 (Figs. 2 and 3). Area 100 contributes the highest proportion of C_{UT} (50.3%) due to high electricity

requirements in the evaporators (EV-101, EV-102, EV-103) used in this stage for the extraction of GO and CPE as well as for the concentration of the free sugars.

Table 3 presents the C_{RM} (M$39.75) employed in the proposed biorefinery. There is no nitrogen source supplementation in the fermentation medium used for succinic acid production because the hydrolysate produced from wine lees is rich in FAN that is sufficient for succinic acid production. The predominant cost of raw materials is attributed to the utilisation of HCl, which is employed in Areas 200, 300 and 400 for the extraction of CTE, pH correction after alkali treatment of lignocellulose-rich solids, extraction of CaT and succinic acid purification.

The succinic acid production stage (Area 400) contributes the highest cost ($M62.9, 43%) in the overall COM (M$145.6) followed by Area 100 focusing on the extraction of free sugars, CPE and GO from grape pomace ($M44, 30%).

3.1.3. *Effect of biorefinery development on the cost-competitiveness of succinic acid production*

The proposed biorefinery using 805,536 t/y winery waste (on wet basis) resulted in the annual production of 30,250 t SA, 8,819 t CPE, 3,763 t GO, 1,982 t CaT and 60,332 t CTE (Fig. 1). Considering the annual succinic acid production (30,250 t/y) and the conversion yield achieved during fermentation (0.64 g/g), the annual sugar requirements is 47,266 t/y. Ioannidou et al. (2020) showed that the aforementioned sugar requirements are available in the winery wastes generated by the predominant wine producing countries in EU, namely Spain (184,000 t sugars/y), Italy (164,000 t sugars/y) and France (151,000 t sugars/y). Based on this estimation, the biorefinery concept presented in this study could be developed in Spain, Italy or France as a central processing facility using waste streams from many wineries. Further process improvement regarding succinic acid production efficiency could reduce further the winery waste requirements.

The main objective of this study was to present the potential of biorefinery development using winery waste on the reduction of the MSP of succinic acid. For this reason, the MSP of succinic acid has been estimated considering winery waste valorisation via either a single-product process or a multiple-product process where a range of market prices for the co-products has been considered. Fig. 4 shows that the annual production of 30,250 t succinic acid using 805,536 t/y winery waste (on a wet basis) without any fractionation (single-product process scenario) leads to a MSP of $4.42/$kg_{SA}$. The waste pretreatment and enzyme hydrolysis efficiency used in the estimation of MSP in the single-product process scenario were the same as the ones achieved in the biorefinery scenario regarding cellulose and hemicellulose to sugar conversion yields (Filippi et al., 2022). This MSP is significantly higher than the current market price of bio-based succinic acid ($2.94/$kg_{SA}$) (E4tech et al., 2015) that is currently used in various applications, ranging from the traditional food and pharmaceutical markets to the production of bio-based polymers and polyester polyols (Ladakis et al., 2018).

The material balances presented in Fig. 1 have been used to estimate the MSP of succinic acid at varying co-product market prices (Fig. 4). The market prices of CPE, GO and CaT have been assumed based on their current market applications. Dimou et al (2016) reported that the

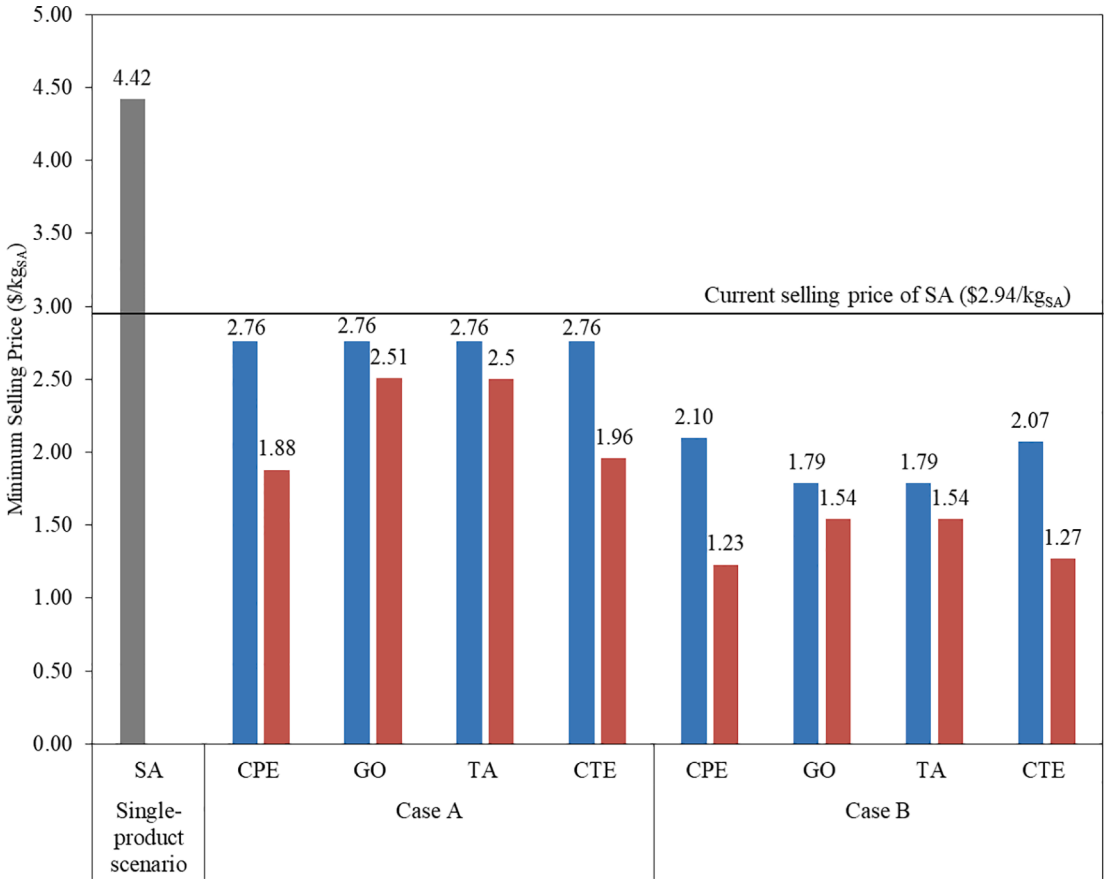

Fig. 4. Estimation of MSP of succinic acid produced from winery wastes via either a single-product process (no biorefinery scenario) or a multiple-product process at varying co-product market prices using a minimum and maximum price range for each co-product. Case A: Blue bars correspond to minimum co-product market prices, while red bars correspond to maximum market price for the main co-product and minimum market prices for the other co-products. Case B: Blue bars correspond to minimum market price for the main co-product and average market prices for the remaining co-products, while red bars correspond to maximum market price for the main co-product and average market prices for the other co-products. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

market prices of antioxidant-rich extracts from grapes may vary within the range of $10–100/kg depending on their purity and the active compounds contained in the extract. In this study, a low CPE market price range ($4–7/kg) has been considered because no further purification has been considered in the performed process design. The GO extracted from grape pomace could be used in culinary, cosmetic and pharmaceutical applications (Jin et al., 2021). In this study, the GO market price range was assumed at $3–5/kg considering that the GO market price may vary within the range of $2–10/kg depending on its final application (Alibaba.com, 2021) and the market price ($4/kg) considered by Jin et al. (2021). CaT is mainly used in the food industry (Dimou et al., 2016). The CaT market price range was assumed at $2–6/kg depending on its final application (Alibaba.com, 2021) and literature-cited data (Dimou et al., 2016).

The CTE extracted in this biorefinery concept as presented by Filippi et al. (2022) could be potentially used in the preparation of bio-based adhesives and resins that are suitable for the production of particleboards in order to substitute for phenol in the production of phenol–formaldehyde resins (Ping et al., 2011). The reagent used for condensed tannin extraction (e.g. NaOH, Na_2CO_3, $NaHCO_3$) and the process used to recover the condensed tannins (e.g. direct lyophilization or HCl treatment after NaOH treatment) affect the properties of the resins and the wood-based panels (Ping et al., 2011; Ping et al., 2012). For instance, the condensed tannins extracted with Na_2CO_3 led to the production of particleboards the properties of which passed relevant international standard specifications for interior grade panels (Ping et al., 2012). It should be pointed out that the adhesive properties of the crude tannin-rich extract extracted in this study has not been verified. Further research is needed to identify the adhesive properties of the tannin-rich extracts produced in this biorefinery concept. In this study, a conceptual approach has been employed to assess the biorefinery development potential if this tannin-rich extract is used for bio-based adhesive preparation. Adjustments in processing conditions and unit operations should be applied for the production of a suitable bio-based adhesive. The CTE market price range ($0.8–1.2/kg) has been assumed considering the market price range of phenol used in the production of phenol–formaldehyde resins (Alibaba.com, 2021).

Fig. 4 presents the MSP of succinic acid at varying co-product market prices using the market price ranges mentioned above. Case A in Fig. 4 presents the MSP of succinic acid at two scenarios where each co-product market price varies between the minimum and maximum price, while the remaining co-products are set at their minimum prices. When the minimum prices of CPE ($4/kg), GO ($3/kg), CaT ($2/kg) and CTE ($0.8/kg) are used, then the MSP of succinic acid acquires the highest value ($2.76/$kg_{SA}$). Even in this extreme case, the MSP of succinic acid is lower than the current market price of bio-based succinic acid ($2.94/$kg_{SA}$). In the case that each co-product market price is set at the maximum price and the remaining co-products are set in their minimum market price, then the most influential co-product is the CPE where a MSP of succinic acid as low as $1.88/$kg_{SA}$ is achieved. Case B in Fig. 4 presents the MSP of succinic acid at two scenarios where each co-product market price varies between the minimum and maximum price, while the remaining co-products are set at their average prices. The CPE is the most influential co-product as the MSP of succinic acid varies from $2.10/$kg_{SA}$ to $1.23/$kg_{SA}$ when the market price of CPE varies from $4/$kg_{CPE}$ to $7/$kg_{CPE}$, while the average market prices of GO ($4/kg), CaT ($4/kg) and CTE ($1/kg) have been used. Fig. 4 shows that biorefinery development can lead to a significantly lower MSP of succinic acid than the current market price of bio-based succinic acid. It should be stressed that if the highest co-product market prices are considered then the MSP of succinic acid is $0.58/$kg_{SA}$.

Fig. 5 shows the variation of NPV as a function of the market price of each co-product considering that the succinic acid market price is equal to the current market price of bio-based succinic acid ($2.94/$kg_{SA}$). The co-product market price range presented above has been used. In each case, the average market price of remaining co-products has been considered. It can be observed that the market prices of CPE and the CTE affect significantly the NPV of the whole biorefinery. In the case of CPE (Fig. 5) this is attributed to their high market price, while in the case of CTE (Fig. 5) this can be attributed to their high production capacity. The estimated DPP ranged from 7 years when the highest market prices of all co-products were considered to 20 years when the lowest co-product market prices were considered. The estimated NPV ranged from $M439.4 when the highest market prices of all co-products were considered to $M39.4 when the lowest co-product market prices were considered.

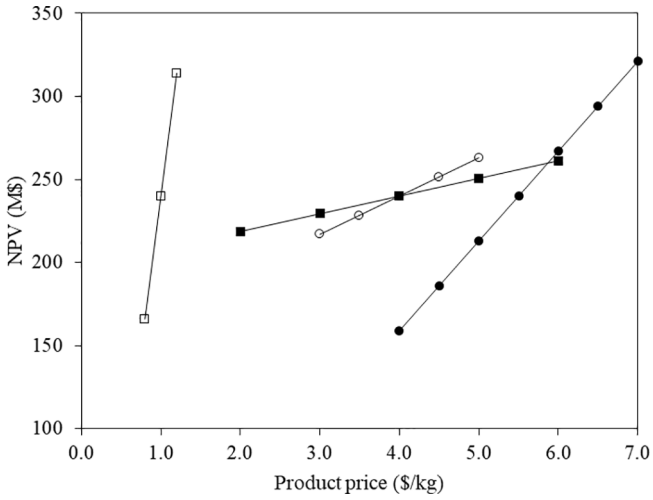

Fig. 5. NPV variation as a function of the individual co-product market price considering CPE (●), GO (○), TA (■), CTE (□). In each case, the average market price of all remaining co-products has been considered, namely CPE ($5.5/kg), GO ($4/kg), CaT ($4/kg) and CTE ($1/kg).

Most literature-cited studies focus on the techno-economic assessment of biorefinery development using individual winery waste streams. Jin et al. (2021) evaluated the profitability potential of three processing scenarios using 33,000 t/y grape pomace for the development of a single-product process producing only grape-seed oil, a two-product process producing grape-seed oil and polyphenols, and a three-product process producing grape-seed oil, polyphenols and biochar. The latter biorefinery scenario was the most profitable one, leading to a NPV of $M111.7 and a payback period of 2.5 years, demonstrating that a multiple-product biorefinery approach ensures process profitability. Dimou et al. (2016) presented a sensitivity analysis based on techno-economic evaluation to assess the development of a profitable wine lees refining process depending on the MSP of the antioxidant-rich extract considering fixed market prices for calcium tartrate ($5/kg), ethanol ($0.6/kg) and yeast cells as animal feed ($1/kg). The COM was estimated at M$1.21 for 500 kg/h wine lees processing corresponding to a MSP of the antioxidants-rich extract of $122/kg. The MSP of the antioxidants-rich extract was reduced to $11.06/kg at 5,000 kg/h of wine lees utilisation. Vega et al. (2021) presented a techno-economic evaluation for polyphenol extraction from red wine pomace via two different extraction methods, solvent extraction and pressurized liquid extraction, in different solvent to dry weight ratios. The processing cost of polyphenol extraction (expressed in kg gallic acid equivalents, GAE) was in the range of €8-26/kg GAE.

3.2. Life cycle assessment

The estimated environmental performance of the selected impact categories (GWP and ADP fossil) of the winery wastes biorefinery is presented in Fig. 6. The FU used is 1 kg total dry waste. Fig. 6A presents the greenhouse gas emissions per FU for the total biorefinery (1.47 kg CO_2-eq/FU) as well as for the individual processing stages (Areas

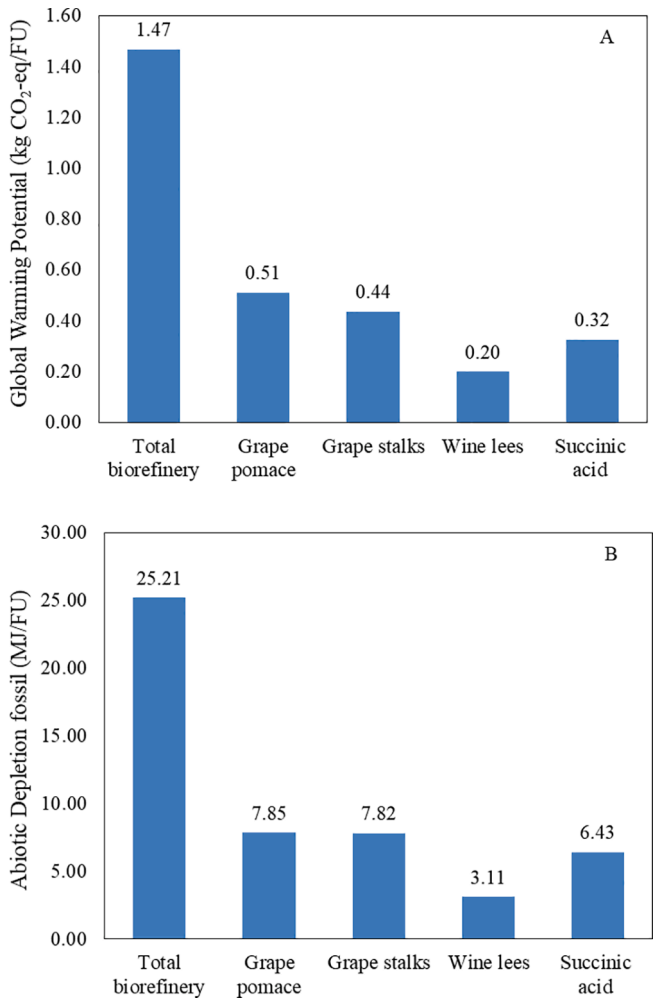

Fig. 6. Global Warming Potential (A) and Abiotic Depletion Potential (B) per kg dry total winery waste expressed for the whole proposed biorefinery and the individual processing stages (Areas 100–400) focusing on grape pomace, grape stalks and wine lees treatment as well as succinic acid production.

100–400 in Figs. 2 and 3). Grape pomace (0.51 kg CO_2-eq/FU) and grape stalks (0.44 kg CO_2-eq /FU) processing contribute the highest GWP including the individual environmental impact of the wastes, namely 2.61×10^{-3} kg CO_2-eq/FU for grape pomace and zero GWP for grape stalks. The ADP fossil for the whole biorefinery (25.21 MJ/FU) is presented in Fig. 6B. The grape pomace (7.85 MJ/FU) and the grape stalks (7.82 MJ/FU) processing stages contribute the highest requirements in non-renewable energy followed by succinic acid production (6.43 MJ/FU) and wine lees processing (3.11 MJ/FU). The environmental impacts of both impact categories are mainly attributed to the utilities consumed for the recovery of the solvents, drying requirements, bioreactor operation and the concentration of free sugars extracted from grape pomace.

Environmental impact assessments of individual winery waste valorisation have been reported in the literature. Cortés et al. (2019) reported the GWP (1.33 kg CO_2-eq/kg wine lees), the ADP (0.435 kg oil-eq/kg wine lees), the terrestrial acidification (4.85×10^{-3} kg SO_2-eq/kg wine lees), and freshwater eutrophication (0.22×10^{-3} kg P-eq/kg wine lees) of wine lees valorisation for the production of antioxidant-rich extract, calcium tartrate and yeast cells. Ncube et al. (2021) presented the environmental impact assessment of conventional wineries integrated with the production of either tartrate or grape-seed oil in order to develop circular patterns. The estimated environmental impact categories for a winery integrated with grape-seed oil extraction from pomace were presented considering 1 bottle of Asprinio wine as functional unit, namely 9.39×10^{-3} kg CO_2-eq for GWP and 2.76×10^{-3} kg oil-eq for fossil resources scarcity (equivalent to ADP fossil). Vega et al. (2021) reported 19 different midpoint indicators for all scenarios assessed for polyphenol extraction from red wine pomace (expressed as kg gallic acid equivalents, GAE). GWP ranged from 27.28 to 171.88 kg CO_2-eq/kg GAE depending on the solvent to dry weight ratio selected for the extraction process, while fossil resources scarcity (equivalent to ADP fossil) ranged from 8.96 to 57.04 kg oil-eq/kg GAE. Ferreira et al. (2018) presented the environmental impact of heat production from grape stalk pellets. Eleven indicators were estimated for the production of 1 MJ heat from grape stalks pellets with GWP and ADP values of 1.45×10^{-2} kg CO_2-eq/MJ and 0.16 MJ/MJ, respectively.

The environmental impact results presented in literature-cited publications cannot be easily compared to the results of this study due to the complexity of implementing the LCA methodology in different biorefineries and the selection of different functional units. The FU selected in this study aimed at evaluating the environmental impact per kg dry waste in order to allow the future comparison of different biorefinery concepts with the valorisation potential of the same waste resource.

4. Conclusions

The profitability potential and environmental impact of a biorefinery using winery waste for the production of succinic acid and various co-products has been presented. The development of marketable co-products is critical in order to achieve process sustainability. Future studies should focus on the development of specific end-products from each extracted fraction with specific market applications. In this way, biorefinery scenarios will be assessed in more detail providing more accurate estimation on process profitability and environmental impact. Furthermore, improving the fermentation efficiency for succinic acid production is also important in order to reduce raw material requirements and production costs.

CRediT authorship contribution statement

Sofia Maria Ioannidou: Conceptualization, Investigation, Software, Writing – original draft, Writing – review & editing. **Katiana Filippi:** Conceptualization, Writing – original draft. **Ioannis K. Kookos:** Data curation, Supervision. **Apostolis Koutinas:** Conceptualization, Resources, Data curation, Supervision, Writing – review & editing. **Dimitrios Ladakis:** Conceptualization, Data curation, Supervision, Writing – review & editing.

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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