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Life Cycle Assessment of Fermentative Production of Lactic acid from Bread Waste based on Process Modelling using Pinch Technology

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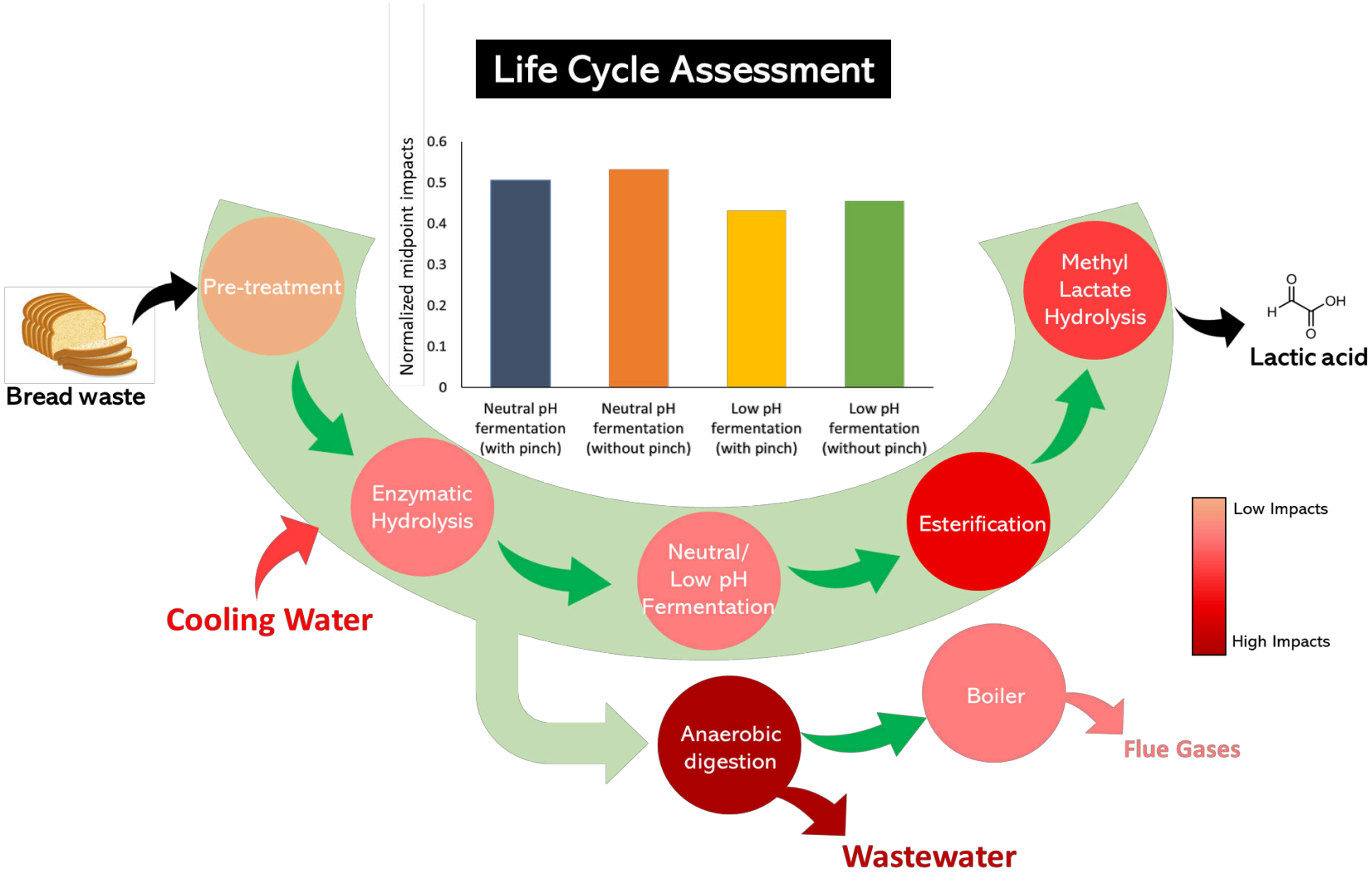
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Graphical abstract



Spotlights

- Bread waste(BW), one of the major sources of global food waste can be a sustainable feedstock for Lactic acid production.
- Relative environmental performance of fermentative lactic acid production technologies from BW was assessed using LCA.
- Low pH fermentation with pinch technology is more environmentally sustainable compared to neutral pH fermentation.
- Wastewater generation from anaerobic digestion and cooling water utility in esterification are environmental hotspots.
- This targeted environmental optimization study will aid policymakers in better decision-making in lactic acid production.

Abstract

Bread waste (BW), which is a rich source of fermentable carbohydrates, has the potential to be a sustainable feedstock for the production of Lactic acid (LA). In our previous work, LA concentration of 155.4 g L^{-1} was produced from BW via enzymatic hydrolysis, which was followed by techno-economic analysis of the bioprocess. This work evaluates the relative environmental performance of two scenarios - neutral and low pH fermentation processes for LA production from BW using a cradle to gate life cycle assessment (LCA). This LCA was based on an industrial-scale biorefinery process handling 100 metric tons BW per day modelled using ASPEN plus. The LCA results depicted that wastewater generation from anaerobic digestion (AD) (42.3 – 51%) and cooling water utility (34.6 – 39.5%) majorly from esterification are the critical environmental hotspots for LA production. Low pH fermentation yielded the best results compared to neutral pH fermentation, with a 11.4 – 11.5% reduction in the overall environmental footprint. Moreover, integration of pinch technology which enhances thermal efficiency and heat recovery within the process led to further reduction in the impacts by 7.2 – 7.34%. Scenario and sensitivity analyses depicted that substituting ultrapure water with completely softened water and sustainable management of AD wastewater can further improve the environmental performance of the processes.

K**eywords:** *Lactic acid; Bread waste; Life cycle assessment; Sensitivity; Uncertainty; Pinch technology*

1. Introduction

Bread waste (BW) is a significant contributor to the global food waste problem, especially due to its shorter shelf life and overproduction, generating an estimated annual waste of 0.9 million tons (Mt), which is approximately 10% of the total production (Kumar et al., 2023). The problem is more predominant in North American and European countries, where bread is a dietary staple. For instance, UK, and Sweden accounted for annual BW production of 0.292 Mt (Narisetty et al., 2021), 0.08 Mt (Brancoli et al., 2019), respectively. The BW proportion in food waste for other European countries

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ranged from 7.9% (Portugal), 13% (Finland), 22% (Netherlands), and 27% (Norway) (Jung et al., 2022). The BW results in significant economic loss along with environmental and social impacts from a life cycle assessment perspective (Brancoli et al., 2020; Narisetty et al., 2021).

In the current times when we are witnessing a push for transitioning from fossil-based economy towards circular bioeconomy (European Commission, 2015), BW has harnessed considerable scientific attention as a valuable renewable feedstock for the production of industrially relevant bio-based materials, such as biochemicals and biofuels. Especially, BW is a clean source of high-quality fermentable sugars, negating the need for physical or chemical pre-treatments that are often required for lignocellulosic feedstocks (Narisetty et al., 2021). Previous studies have demonstrated the recovery of several value-added organic chemicals from BW including succinic acid (Leung et al., 2012), ethanol (Narisetty et al., 2022a; Pietrzak and Kawa-Rygielska, 2014), lipids (Thyagarajan et al., 2014), enzymes (Haque et al., 2016), biohydrogen (Han et al., 2017), 2,3-butanediol (Narisetty et al., 2022b), etc., One of the most promising biochemicals that can be produced from BW is lactic acid (LA). According to US Department of Energy, LA is a top platform chemical (Bozell and Petersen, 2010), which has numerous applications in various sectors, including pharmaceuticals, cosmetics, food, specialty chemicals, textiles, and leather (Kim et al., 2022). Especially, with the growing demand for polylactic acid (PLA) based bioplastics, LA production is estimated to reach 1,960 kilotons by 2025 (Cox et al., 2022) from reported 1220 kiloton in 2016 (Abedi and Hashemi, 2020).

Unlike chemical routes of LA synthesis, which are expensive, energy-intensive, fossil fuel dependent, and can release hazardous by-products, the microbial fermentation route of LA production is environmentally and economically more sustainable (Kim et al., 2022). Previous studies have demonstrated the potential of LA production from BW through microbial fermentation mostly using *B. coagulans* for fermentation. The productivities of LA in these studies varied 2.59 g/(L.h) (batch mode), 11.28 g/(L.h) (continuous mode) (Alexandri et al., 2020), 2.67 g/L.h (Olszewska-Widdrat et al., 2020). In our previous work, high-level LA production (titre: 155.4 g/L; yield: 0.85 g/g; productivity: 1.30 g/L. h)

was achieved using glucose rich hydrolysate from enzymatic saccharification of BW by *B. coagulans* (Cox et al., 2022). Further, this process were modelled using Aspen Plus software to simulate a centralized biorefinery with 100 metric ton per day (MT/day) BW handling capacity which was evaluated using techno-economic and profitability analyses (Mailaram et al., 2023). Although the prospective industrial relevance of BW- derived LA production through enzymatic fermentation is evident, the environmental impacts associated with these processes are critical in determining the sustainability of the technology. Life cycle assessment (LCA) is a common tool used to determine the environmental impact of the processes. Some of the LCA studies on valorisation of BW through biorefinery approaches, such as for bioethanol production (Brancoli et al., 2020; Narisetty et al., 2022a), succinic acid recovery (Gadkari et al., 2021), recovery of microbial protein, LA, succinic acid from mixed municipal solid waste (Khoshnevisan et al., 2020) have yielded positive results. Li et al., (2021), on the other hand, carried out techno-economic and life cycle assessment to compare the efficacies of neutral and low-pH fermentation processes for lignocellulosic biomass to LA conversion.

Microbial fermentation is generally carried out using *Bacillus coagulans* DSM1 (neutral pH fermentation), in which neutral pH is maintained using calcium hydroxide. The calcium lactate is subsequently hydrolyzed by sulphuric acid with the formation of calcium sulfate. Low pH fermentation using acid-resistant engineered yeast is a potential alternative to circumvent process complexity and disposal of inorganic salt. The Cargill has been commercially producing LA by low pH fermentation, with a LA titer of 130 g/L and productivity of 3 g/(L.h) (Marchesan et al., 2021). However, there is a lack of comprehensive LCA studies to assess the relative environmental sustainability of BW to LA production technologies.

The objective of the current study is to investigate the relative environmental performance of neutral pH fermentation and low pH fermentation processes for LA production with BW as the feedstock. The reliable material and energy balance data is critical in clearly identifying the environmental impacts of these processes. In this work, these data were generated from our

comprehensive process design based on experimental data and validated kinetic/thermodynamic models using Aspen Plus (Mailaram et al., 2023). Following the principles of circular biorefinery, the process of BW to LA fermentation was also integrated with anaerobic digestion (AD) of the residual BW for biogas recovery and use for energy generation. The processes were further integrated by pinch technology, which is a crucial tool for improving the thermal efficiency of the process by maximizing the process heat exchange through cooling water recirculation. Using a cradle-to-gate LCA, the study aims to determine the relative environmental impacts of processes of LA production and to identify the critical hotspots and categories of maximum concern. In addition, the study aims to assess the extent of environmental impact reduction achieved through the integration of pinch technology compared to without pinch technology. Moreover, sensitivity and scenario analyses were also performed on the identified hotspot parameters to analyse the stability of these impacts with variability in the parameters and their values. In addition, an uncertainty analysis was also performed to evaluate the effects of uncertainty on the original data on the variability of the associated environmental impacts. We envision that this novel study will aid policy-makers and LCA practitioners in better decision-making regarding LA production technologies from BW through a comprehensive view of their environmental assessment.

2. Process description

The LCA study is based on an industrial-scale LA production biorefinery with 100 MT/day BW processing capacity based on the models developed in our previous techno-economic analysis (Mailaram et al., 2023) and the experimental data as provided by our lab-scale experiments (Cox et al., 2022). Process simulation includes all data estimation methods that involve the use of ASPEN PLUS and its databases. Lab-scale operation parameters in BW to lactic acid study were used as inputs for the simulation as a result of which energy flows, material flows, and elementary flows were obtained. The study encompasses two scenarios following enzymatic hydrolysis: (1) neutral pH fermentation using *Bacillus coagulans* which is a homofermentative bacteria that functions at neutral pH and (2) low pH fermentation, which employs acid-resistant engineered yeast for fermentation. Moreover, the study

also evaluates the relative performance of two process models: 'Without Pinch' and 'With Pinch technology'. The later entails an optimal design of heat exchanger network (HEN) to achieve maximum process heat energy utilization. The process model was divided into five sub-processes: pre-treatment, hydrolysis, fermentation, downstream separation processes including esterification and methanol hydrolysis, and other auxiliary processes. The pre-treatment phase involves blending shredded pieces of BW with water to form a slurry of 20 % w/v solid loading rate, which was sterilized using autoclave (121 °C for 15 min). Further, the autoclaved slurry was treated with an enzyme (Dextrozyme) in a carbohydrate hydrolysis reactor. The resulting sugar solution from the saccharification reactor was then sent to a fermenter after the removal of unconverted solids.

In Scenario 1, the reactive separation method was used to recover LA from fermentation broth. In this process, calcium hydroxide was added to maintain neutral pH in the fermenter by converting LA to calcium lactate, which was subsequently hydrolyzed to LA by sulfuric acid. Calcium sulphate - the by-product of the process. was separated by centrifugation, and the liquid stream containing LA, unconverted sugars, protein, fats, and salts was sent to a distillation column (DC1) to remove around 99% water as distillate. The water was recycled to the saccharification process after purging the excess amount. The concentrated LA obtained from the bottom of DC1 was mixed with methanol after cooling and sent to the LA esterification reactor. Methyl lactate and the excess methanol were recovered as distillates after separation from unconverted LA and heavy residues by two different distillation columns (DC2 and DC3). These heavy residues, along with solid residues from the saccharification process were sent to AD, while the methanol was recycled to the LA esterification reactor. Methyl lactate was hydrolyzed (over cation exchange resin catalyst) in DC4-DC6 to obtain methanol, water, LA, and unconverted methyl lactate. The recovered LA undergoes purification by vacuum distillation.

In Scenario 2, where acid-resistant microorganisms were used for fermentation, the predominant portion of LA was present in acid form unlike scenario 1. So, the steps of lime addition for acid neutralisation, conversion to calcium lactate, separation of calcium sulphate, and subsequent hydrolysis

of calcium lactate to LA were redundant. In this scenario, the fermentation broth containing LA was directly sent to DC1, and the remaining process was the same as in Scenario 1. Fig. 1 depicts the biochemical pathway for LA production from glucose as a carbon source by *Bacillus coagulans*. All the solid residues and liquid effluents from hydrolysis, fermentation, and esterification processes were aggregated and subjected to anaerobic digestion to generate biogas. This biogas was channelled to a boiler unit to generate high-pressure steam which was used to complement energy demands partly. Detailed flowsheet depicting the individual processes modelled for scenarios 1 and 2 were shown in Fig S1 and S2.

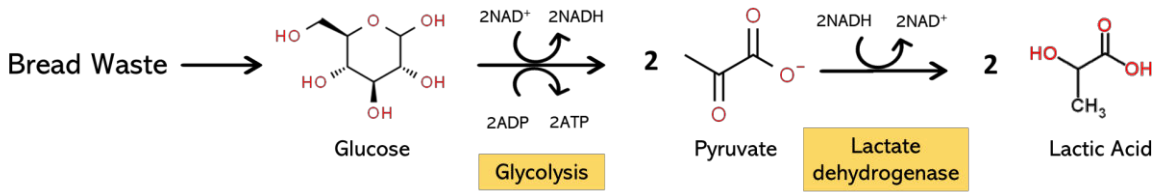


Fig. 1. Biochemical pathway from glucose to lactic acid (LA) production

3. LCA methodology

3.1 Goal and scope

The goal of this study is to evaluate and compare the environmental footprints of two pathways of fermentative LA production from BW to determine their relative environmental performance. A cradle-to-gate type of system boundary was considered for this study (Fig. 2), and 1 kg of LA production was chosen as the functional unit. Considering LA as a chemical intermediate with extensive applications beyond its current use, its product logistics, use, and end-of-life considerations are deemed as out of the scope of the current study. The geographical scope of this study was limited to the United Kingdom (UK) wherein the lab-scale experiments were performed.

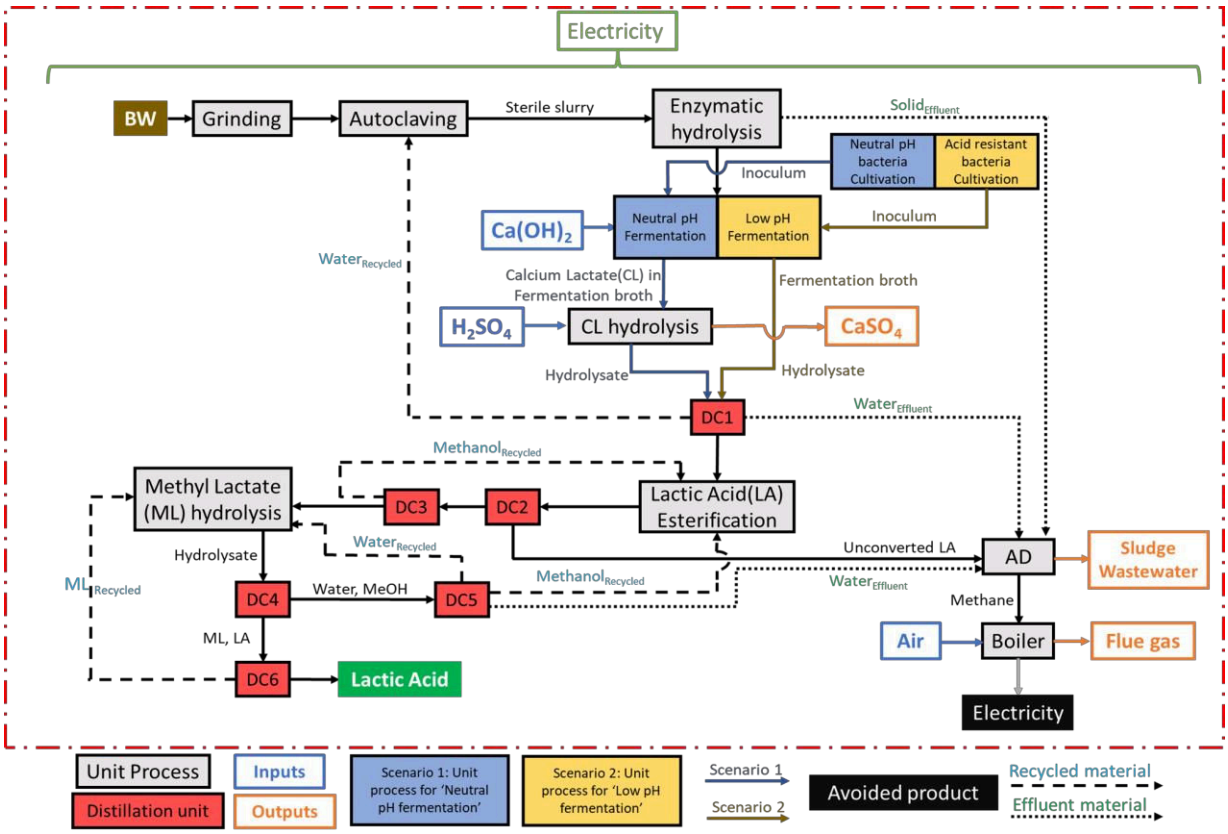


Fig. 1: System boundary for LA production from BW via neutral pH fermentation (Scenario 1) and low pH fermentation (Scenario 2); DC-Distillation column, CL-Calcium lactate, AD-Anaerobic digestion

3.2 Life cycle inventory (LCI)

The LCI for the industrial scale LA production from BW with a handling capacity of 100 MT/d through neutral pH fermentation and low pH fermentation are presented in Table . The inoculum preparation phase was adopted from our previous work (Cox et al., 2022). The process inventory depicting the main input/output data for various stages of the biorefinery were extracted from simulations developed in Aspen Plus (Mailaram et al., 2023). The background footprint data of the products and processes in LCI were obtained from EcoInvent 3.8 database (Table S1). It should be noted that the energy requirements of pre-treatment process depicted in the LCI included crushing and autoclaving for simplification. Similarly, the chemical requirements in the cultivation phase were also included in the fermentation process. All the electricity requirements of small sub-processes such as centrifugation, pumping, distillation, etc., were included in their individual processes.

The list of assumptions considered while compiling the LCI are as follows:

- Density of the hydrolysate generated after enzymatic hydrolysis was assumed to be 1.1 kg L^{-3} (Tiwari et al., 2023).
- As KH_2PO_4 was not available in the EcoInvent 3.8 database, its consumption was calculated based on the stoichiometric requirement of H_3PO_4 & KOH.
- As $MgCl_2$ was not available in the EcoInvent 3.8 database, its consumption was calculated based on the stoichiometric requirement of MgO, H_2O & HCl.
- As $Na_2PO_4.6H_2O$ was not available in the EcoInvent 3.8 database, its consumption was calculated based on the stoichiometric requirement of Na_2HPO_4.
- Inoculum preparation was assumed to be done at neutral pH.
- The amount of wastewater to be treated was calculated by assuming its density to be 1 kg/m^3 (Rhee et al., 2021). Chemical oxygen demand (COD) of influent to AD is assumed to be 5 g/L (Wang et al., 2003), with COD removal efficiency being assumed as 70%. The wastewater volume generated from AD was represented as an equivalent to the average wastewater (COD = 0.16 g/L), available in the EcoInvent database.
- As the plant location is in the UK, all the data including electricity production mix, chemicals, and any other secondary data input into the LCA was based on the UK or European averages available in the database.
- The environmental impacts associated with the infrastructure manufacturing, maintenance and land area usage were ignored as it is assumed to be identical for all the scenarios considered in the study. The present study has excluded the impact associated with the capital goods and infrastructure due to the absence of relevant datasets in the EcoInvent 3.8 database. Similarly, other LCA studies on biorefineries have not included capital goods due to the same reason (Ebrahimian and Mohammadi, 2023; Kiehbadroudinezhad et al., 2023; López-Herrada et al., 2023). Moreover, similar approach has also been adopted by other studies which have focused only on the material and energy balances based on a technoeconomic assessment modelled in

ASPEN PLUS (Jasper et al., 2022; Liu et al., 2021; Tiwari et al., 2023; Vinardell et al., 2023). Previous studies have also depicted negligible environmental effects of infrastructure for large-scale processes with very long lifespan as a reason for their exclusion (Bhar et al., 2022; Li et al., 2022; Smidt et al., 2015).

- For the purpose of simplification, BW was assumed as post-consumer waste, and so all the environmental impacts associated with its product lifecycle till waste generation were excluded from the analysis.
- The emissions associated with the collection, separation, and transportation of the BW and supplementary chemicals to the biorefinery were assumed to be negligible and so not considered in the current study. Similar studies in the literature have also assumed that the impacts of segregation (Elginoz et al., 2020) and transportation are negligible (Brunklaus et al., 2018; Cok et al., 2014; Gadkari et al., 2021; Moussa et al., 2016).

Table 1: Inventory data for LA production from 4166.7 kg of BW by neutral pH fermentation (Scenario 1) and low pH fermentation (Scenario 2).

			INPUT consumed/OUTPUT produced per hr			
			Neutral pH fermentation (Scenario 1)		Low pH fermentation (Scenario 2)	
		Unit	With Pinch	Without Pinch	With Pinch	Without Pinch
Pretreatment	BW (Input)	kg	4,166.7	4,166.7	4,166.7	4,166.7
	Electricity (Input)	kWh	145.6	145.6	145.6	145.6
Hydrolysis	Electricity (Input)	kWh	2.24	2.24	2.24	2.24
	Steam (Input)	kWh	249.7	669	249.7	669
	Cooling Water (Input)	kg	73,150.42	326,325.4	73,150.4	73,150.4
	Enzyme (dextrozyme) (Input)	kg	99.53	99.53	99.53	99.53
	Hydrolysate (Output)	kg	18,156.02	18,156.02	18,156.02	18,156.02
Fermentation	Electricity (Input)	kWh	14.17	14.17	1.49	1.49
	NH_2SO_4 (Input)	g	1,625.79	1,625.79	1,625.79	1,625.79
	KH_2PO_4 (Input)	g	214.57	214.57	214.57	214.57
	$CaCl_2$ (Input)	g	20.63	20.63	20.63	20.63
	$MgCl_2$ (Input)	g	165.05	165.05	165.05	165.05
	$Na_2PO_4.6H_2O$ (Input)	g	1,790.84	1,790.84	1,790.84	1,790.84
	Yeast extract (Input)	g	1,650.55	1,650.55	1,650.55	1,650.55
	$Ca(OH)_2$ (Input)	kg	806.3	806.3	0	0

	H_2SO_4 (Input)	kg	1067.3	1067.3	0	0
	Cooling Water (Input)	kg	178,852.6	178,852.6	115,246.2	115,246.2
	Steam (Input)	kWh	0	377.8	0	0
	Fermentation Broth (Output)	kg	18540.4	18540.4	18,156.02	18,156.02
	$CaSO_4$ (Output)	kg	1481.5	1481.5	0	0
Downstream separation processes						
Esterification	Electricity (Input)	kWh	23.68	23.68	6.72	6.72
	Cooling Water (Input)	kg	2,338,040	2,566,837	2,365,281	2,522,962
	Steam (Input)	kWh	13,743.1	15,070.9	13,494.6	14,802.1
ML hydrolysis	Electricity (Input)	kWh	1.55	5.14	1.55	1.55
	Steam (Input)	kWh	2,160.9	2,451.7	2,172.8	2,444.8
	Cooling water (Input)	kg	422,988.9	455,672.1	422,122.8	454,705.9
	Lactic acid (Output)	kg	1,629.34	1,629.34	1,629.34	1,629.34
Auxiliary units						
Anaerobic digestion	Cooling Water (Input)	kg	24,073.2	24,073.2	19,077.3	19,077.3
	Steam (Input)	kWh	0	440.5	0	440.9
	Wastewater (Output)	kg	42,396.48	42,396.48	33,569.29	33,569.29
Boiler	Electricity (Input)	kWh	186.5	186.5	186.5	186.5
	Air (Input)	kg	5,867	5,867	5,867	5,867

	H_2O (Output)	kg	842.6	842.6	841.9	841.9
	CO_2 (Output)	kg	1,441.5	1,441.5	1,438.4	1,438.4
	NO_2 (Output)	kg	169.97	169.97	169.97	169.97
	N_2 (Output)	kg	4,500.5	4,500.5	4500.4	4500.4
	O_2 (Output)	kg	48.35	48.35	49.93	49.93
	Energy generated (Output)	kWh	4,579	2.2	4,579	2.2

3.3 Life cycle impact assessment (LCIA)

The LCA was modelled using SimaPro v9.4.0.2 PhD. ReCiPe 2016 (H) is one of the most commonly used LCIA methodologies and was therefore adopted in this study. It evaluates the environmental footprint at both midpoint and endpoint levels. Among the midpoint impact categories, the present study focuses on global warming potential (GWP), terrestrial acidification potential (TAP), freshwater eutrophication potential (FWEP), marine eutrophication potential (MEP), carcinogenic human toxicity (HTc), non-carcinogenic human toxicity (HTnc), terrestrial ecotoxicity potential (TETP), freshwater ecotoxicity potential (FWETp), and marine ecotoxicity potential (METp). Moreover, based on the recommendations of ISO 14044, the present study adopted an 'avoided burden' and 'system expansion and substitution' approach to allocate the impacts.

3.4 Sensitivity and uncertainty analysis

The overall environmental impact of a process is often dictated by some specific input or output parameters. Therefore, the significance of the parameters was addressed via sensitivity analysis, and the same were varied by ± 5% and ± 10%. Sensitivity analysis highlights how the variations in the selected parameters can change the overall contribution analysis. Additionally, the uncertainty analysis was based on the Monte Carlo method. The same was modelled in the SimaPro software by running 10,000 simulations at 95% confidence interval, and the analysis was performed using ReCiPe 2016 Midpoint (H) method.

4. Results and Discussion

4.1 Life cycle impact assessment

The relative environmental performance of neutral pH and low pH fermentation technologies associated with LA production from BW were analysed at the midpoint and endpoint levels (Fig. 3). Also, detailed midpoint characterization data for all the technologies with individual contributions of each sub-process were also presented in the supplementary information (Table S2-S5). Moreover, their variation in environmental impacts with and without pinch technologies was also evaluated. The

adoption of pinch technologies maximized heat exchange through in-process cooling water recirculation, while running LCA without pinch technologies did not host such mechanisms. In all these processes, the relative importance of each of the impact categories was evaluated, and the categories of significant concern were identified. Moreover, the hotspot sub-processes and pathways, which are expected to be of particular interest to the LCA practitioners and policy-makers were recognized, and their environmental implications were critically discussed in this section. It may be noted that the normalized scores represented in both midpoint and endpoint characterisation analyses represent the environmental impacts associated with one kilogram of LA production from BW.

The midpoint normalization results depicted that freshwater eutrophication, human carcinogenic toxicity and marine water eutrophication were the major environmental impact categories for both technologies. Their respective contributions ranged from 0.12 to 0.145 kg P eq, 1.02 – 1.28 kg 1,4-dichlorobenzene (DCB)-eq, and 0.133 to 0.166 kg N eq, accounting for approximately 49 – 54%, 26.2 – 30.1%, and 7.7 – 8.7% of the total negative impacts. These impacts can be majorly attributed to the AD and esterification processes, which accounted for 42.3 – 51%, and 34.6 – 39.5% of the overall impacts of LA production from BW. However, it should be noted that the positive environmental impacts of water consumption impact category majorly contributed by reduction of water consumption in AD process were able to negate 15.9 – 19.2% of the overall negative impacts resulting in significant reduction in the total ecological footprint. Eutrophication was also depicted as an important environmental impact category in previous LCA studies on LA production (Marchesan et al., 2021; Ögmundarson et al., 2020). So, AD and esterification processes can be considered major environmental hotspots during the production of LA from BW. The effects of freshwater ecotoxicity (0.22 – 0.276 kg 1,4-dichlorobenzene (DCB)-eq) and marine ecotoxicity (0.291 – 0.367 kg 1,4-dichlorobenzene (DCB)-eq) along with ozone formation on human health (0.14 – 0.15 kg NOx eq) and on terrestrial ecosystems (0.141 – 0.151 kg NOx eq) were other impact categories of significance contributing 1.03 – 3.29%, 0.48 – 2.53%, 2.03 – 2.31% and 2.36 – 2.68% to the total impacts, respectively.

The boiler process (69.3 – 73.6%) was the major factor responsible for the effects of ozone formation; whereas AD (90.6 – 96%) came out to be the biggest contributor to the effects of ecotoxicity. On the other hand, global warming, terrestrial acidification, fossil resource scarcity, and human non carcinogenic toxicity were the other minor factors accounting for 3.3 – 3.5% of the total impacts. Although previous literature has highlighted the prevalence of these impact categories (Table 2), it is important to note that these studies have taken into account the production pathways of renewable feedstocks and LA downstream purification processes in their LCA, which significantly contributed to the observed impacts (Harbec, 2010; Mandegari et al., 2018; Ögmundarson et al., 2020). Other processes, including methanol hydrolysis (6.2 – 7%), boiler (3.4 – 3.92%), fermentation (1.74 – 2.76%), and hydrolysis (1.9 – 5.84%), also had minute effects on the overall midpoint impacts.

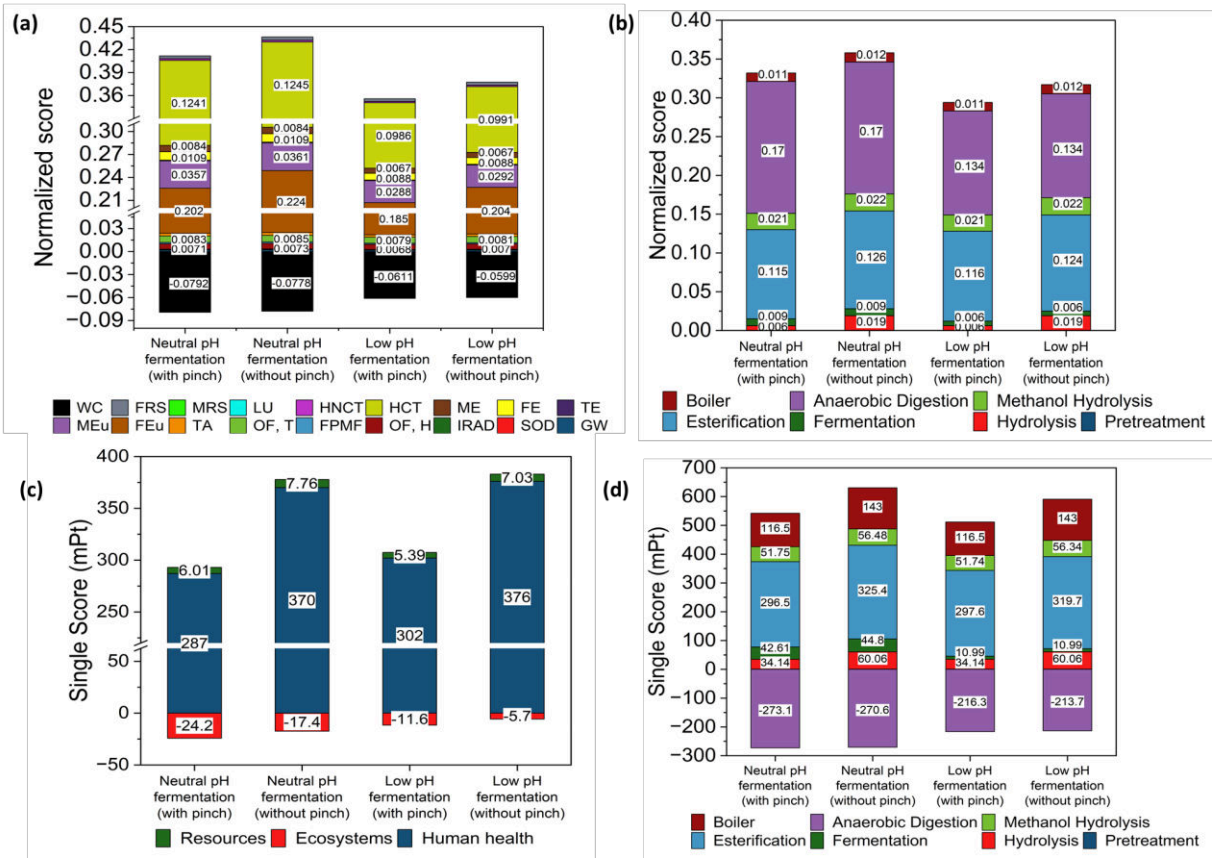


Fig. 3: Comparative midpoint and endpoint normalization data for each scenario. (a) classification based on midpoint impact categories; (b) classification of midpoint impacts based on individual processes; (c) classification based on endpoint damage categories; (d) classification of endpoint

impacts based on individual processes; WC: Water consumption; FRS: Fossil resource scarcity; MRS: Mineral resource scarcity; LU: Land use; HNCT: Human non-carcinogenic toxicity; HCT: Human carcinogenic toxicity; ME: Marine ecotoxicity; FE: Freshwater ecotoxicity; TE: Terrestrial ecotoxicity; MEu: Marine eutrophication; FEu: Freshwater eutrophication; TA: Terrestrial acidification; OF, T: Ozone formation, Terrestrial ecosystems; FPMF: Fine particulate matter formation; OF, H: Ozone formation, Human health; IRAD: Ionizing radiation; SOD: Stratospheric ozone depletion; GW: Global warming;

Considering the endpoint damage categories, resources was the most prominent damage category contributing to 0.755 – 1.09 USD2013, which accounts for most of the total endpoint negative impacts. Esterification and AD were the major processes contributing up to 55.7 – 72.4%, and 19.4 – 26.8% of the total impacts of resources. Esterification process also had a major negative environmental influence on the other two endpoint damage categories of human health and ecosystem contributing up to 51 – 65 % of the total negative impacts. However, AD process was able to negate 30.2 – 42.1 % (human health), and 112 – 158 % (ecosystems) of the total negative endpoint impacts. The reduction of midpoint impacts in the water consumption category corresponds to the decrease in endpoint impacts with increase in the wastewater generation during AD. One of the plausible reasons behind this observation can be the comparatively higher weightage of midpoint to endpoint conversion factors of water consumption as compared to other midpoint impact categories. Another reason can be the release of 0.9 m^3 of treated water back into the environment per m^3 of treated wastewater as documented in the EcoInvent 3.8 database, which translates to reduced water consumption. According to the ReCiPe 2016 methodology, water consumption can be translated to endpoint impacts of human health and ecosystem. While the water stress index (WSI) (i.e., 0.657) and potential of malnutrition due to lack of water transfer to endpoint category of human health, Net primary productivity (NPP) is used to convert the water consumption impacts to the endpoint impact on ecosystems. So, the negative end point impacts in human health damage category varied from 1.72 – $2.25*10^{-5}$ DALY, while the ecosystems category contributed to positive endpoint impacts varying from 2.1 – $8.94 *10^{-8}$ species.yr.

4.2 Detailed contribution analysis

Considering the individual processes, it can be deduced that AD was the hotspot with very high environmental impacts. The environmental footprint of AD was majorly driven by wastewater generation from the process, which was responsible for impacts in fresh and marine water eutrophication categories. This can be due to the assumption that wastewater from AD was released directly into the environment without any treatment in this LCA study. The massive wastewater generation from biorefinery processes for the production of LA was also depicted by previous studies as a major disadvantage in the economic and environmental sustainability of the process (Gezae Daful and Görgens, 2017; Liu et al., 2021, 2015). The environmental impacts of esterification, on the other hand, were majorly contributed by the very high utility of cooling water in the process, which accounted for almost 97% of its impacts. Cooling water utility was also the major factor in other processes, including methanol hydrolysis (97.6 - 97.7%), fermentation (93 – 100%), and hydrolysis (56 - 84%), making it another critical hotspot in the LA production. The environmental significance of wastewater generated from AD and the water utility in the processes have also been demonstrated by other studies (Harbec, 2010; Khoshnevisan et al., 2020; Ögmundarson et al., 2020).

The boiler process, which is responsible for 3.4 – 3.9% of the effects, was majorly associated with the gaseous emissions from the burning of methane generated from the AD process. However, the energy generation from methane combustion in with pinch scenarios was able to negate up to 8.93% of the environmental impacts of boiler process, which was not the case without pinch technologies. Although the effect of the use of chemicals (7%) and enzymes in the process of hydrolysis (15 - 44%) in neutral pH fermentation process were important contributors within those processes, their overall environmental impact on the entire LA production was fairly insignificant. Similarly, the chemicals used in the inoculum cultivation contributed to 0.35 – 0.59 %, and 0.69 – 2.74 % of the mid and end point impacts of the fermentation process. However, its contribution in the overall process varied from 0.01% (midpoint) and 0.1% (endpoint) impacts of the total process, which was very insignificant. In contrast,

some of the studies (Gezae Daful and Görgens, 2017; Li et al., 2021; Mandegari et al., 2018) have depicted higher environmental sensitivity of their processes to the use of chemicals. This could be due to the use of different LCA methodologies, where previous studies have prioritized assessing global warming potential. Also, the use of electricity was an integral part of most of the processes; however, its influence was always less than 1% of the total impacts within the individual processes.

Comparing the individual technologies, it is clear that low pH fermentation exhibited a relatively lower environmental footprint (by 11.4 – 11.5%) compared to the neutral pH fermentation. This variation is majorly due to the reduced prominence of AD and its associated impact categories in the low pH fermentation technology. Especially the lower wastewater generation rate by almost 21% was the major factor responsible for the same. Additionally, the use of chemicals (lime, sulphuric acid, and gypsum) for pH neutralisation also increased the environmental impacts of the neutral pH fermentation process by a very insignificant amount. Similarly, Li et al., (2021) also reported lower environmental impacts by 15%, 5% in the categories of 100-year global warming potential and fossil energy consumption in low-pH fermentation technology for LA production from lignocellulosic materials as compared to the neutral pH fermentation route.

On the other hand, comparing with and without pinch technologies, with pinch technologies were environmentally superior by 7.2 – 7.34% compared to the later. This variation within impacts was majorly contributed by the hydrolysis and esterification processes which were responsible for 47 – 54%, 34 – 43% (midpoint), and 29 – 32%, 27 – 32% (endpoint) of the total impact differences, respectively. This was due to the high cooling water usage of without pinch technologies, which would evidently be reduced in with pinch technologies due to recirculation for heat exchange within the system. The cooling water recirculation and its associated reduction in environmental midpoint impacts could also be reflected in the other processes, such as boiler (by 8.3%) and methanol hydrolysis (by 8.7%) in pinch technology. Mussatto et al., (2013) and Li et al., (2021) also adopted energy integration with heat exchangers in their LA production process for higher environmental performance.

It should also be noted that the heat recovery equipped with pinch technology at different stages of the process has also substantially reduced the steam requirement as compared to without pinch technologies. Especially the combined steam requirement in AD and esterification processes was reduced by 6,366 MJ with the integration of complete heat recovery and re-utility in pinch technologies which was responsible for reduction in 0.5% of their midpoint environmental impacts. Similarly, the heat recovery in neutral pH fermentation helped in the reduction of steam requirement by about 1,360 MJ, which in turn reduced the environmental impacts of the individual process by 1%. There was a significant reduction in steam consumption in other stages, such as hydrolysis (by 63%), and methanol hydrolysis (by 8.8%). However, due to the very low environmental footprint of steam, it has not contributed to any significant differences in environmental impacts between with and without pinch processes. Mussatto et al., (2013) in their LCA study also reported a higher environmental influence of mass integration on water (including water treatment) compared to energy integration with heat exchangers. But, the study suggested combining energy and mass integration for the best environmental performance of a biorefinery, which is adopted in the current study.

So, it can be deduced that low pH fermentation with pinch technology was the most environmentally sustainable out of all the compared technologies. Further, wastewater generation from AD and cooling water utility from esterification, hydrolysis, and methanol hydrolysis were the definitive hotspots identified, which should be of particular interest while designing the systems. Overall, focussing on environmental optimization of the process in ways to recycle the wastewater and reduce the use of cooling water can help in making the process greener and more sustainable.

Table 2: Comparison with other LCA studies on fermentative LA production from different renewable feedstocks

S.No	Feedstock	Fermentation method	LCA Methods	Environmental Hotspots	Major environmental impact categories	Reference
1.	Lignocellulosic feedstock	Neutral and low-pH fermentation	LCA in Python using the simulated stream (input chemicals and emissions) and utility inventories from BioSTEAM.	• Natural gas and electricity used in boiler • Lime used in fermentation process	• 100-year global warming potential (GWP_{100}) • Fossil energy consumption (FEC)	(Li et al., 2021)
2.	Sugarcane bagasse and leaves	Neutral and low-pH fermentation	ReCiPe Midpoint(H)	• Use of $Ca(OH)_2$ and H_2SO_4 and production of gypsum ($CaSO_4$) as solid waste in neutral pH fermentation • Use of nutrient for the strains in low pH fermentation	• Land occupation • Human Toxicity • Marine aquatic ecotoxicity • Global warming • Abiotic depletion	(Gezae Daful and Görgens, 2017)
3.	Municipal Solid Waste	Electrodialysis of fermentation broth using ion-exchange membranes	Impact 2002+	• Anaerobic digestion process • Combined heat and power production	• Climate change • Human health damage	(Khoshnevisan et al., 2020)
4.	• Corn • Corn stover • *Laminaria sp.*	Neutral pH fermentation	Recipe Midpoint(H), Recipe Endpoint (H)	• Energy utility • Use of triethanolamine in LA purification • Treatment of refinery sludge	• Global warming • Ozone formation • Fine particulate matter impacts • Terrestrial acidification • Terrestrial ecotoxicity • Mineral resource scarcity • Fossil resources scarcity • Freshwater and	(Ögmundarson et al., 2020)

					marine water eutrophication	
5.	Sugarcane bagasse	Neutral pH fermentation	CML-IA baseline 3.02 model	• Chemical consumption • Emissions to the atmosphere from LA purification stage	• GWP_{100} • Eutrophication • Acidfication	(Mandegari et al., 2018)
6.	Potato waste starch	Neutral pH fermentation	IMPACT 2002+	• Water utility • Saturated steam in LA purification stage	• Resource depletion • Climate change	(Harbec, 2010)
7.	Bread waste	Neutral and low-pH fermentation	Recipe Midpoint(H), Recipe Endpoint (H)	• Wastewater generation from Anaerobic digestion • Cooling water in Eutrophication	• Freshwater and marine water eutrophication • Human carcinogenic toxicity • Water consumption • Ozone formation	Current study

4.3 Recommendations for reducing the environmental footprint

The present study observed the wastewater generation from AD to be one of the most critical parameters which defines the overall environmental performance of the process. Therefore, the integration of technologies which promote sustainable valorisation of wastewater into value-added products can substantially reduce the environmental impacts associated with its in-process generation (Lamolinara et al., 2022). Considering the wastewater digestate from AD, mostly constituting partially degraded residual organic materials and inorganic contaminants, it can be valorized via nutrient recycling through biochemical or thermo-chemical postprocessing. This can help in minimizing the environmental impacts caused by nutrient pollution, such as eutrophication, soil toxicity, algal blooms, microbial contamination, among other factors (Jurgutis et al., 2021). Previous LCA studies have also reported that the combined emissions of wastewater AD and utilization of the digestate as an organic fertilizer were lower than the production and use of mineral fertilizers (Timonen et al., 2019). Khoshnevisan et al., (2020) reported a significant decrease in the environmental impacts in human health, resource depletion, climate change, and ecosystem damage categories with the additional use of AD digestate as a biofertilizer in agriculture along with LA production. However, it should be noted that wastewater with high water content and low-nutrient concentration might have a higher energy and environmental footprint for use in agriculture (Herrera et al., 2022). In this case, depending on its organic composition, the use of thermochemical technologies, such as hydrothermal carbonization, aqueous phase reforming, supercritical water gasification, or hydrothermal liquefaction, for energy recovery from the wastewater can also be suggested (Sharma et al., 2022). However, the techno-economic feasibilities and environmental impacts associated with integrating these additional processes need to be analysed before suggesting them as a potential solution.

The present study also witnessed a massive environmental footprint due to the cooling water utility. Such high water consumption has been observed to be associated with fermentative LA production processes from different feedstocks (Ma et al., 2022; Wang et al., 2021). The water has been typically

used in acid, alkali, or solvent solutions, or used for washing of feedstock to decontaminate along with its use as steam condensation water (Liu et al., 2015). To reduce the overall water consumption in the process, wastewater recycling after treatment was proposed as a sustainable practice by some of the previous studies. For instance, in a similar study of neutral and low pH fermentation for LA production, Li et al., (2021) integrated AD for wastewater treatment with sludge dewatering, membrane bioreactor, and reverse osmosis for water recovery. This water was recirculated in the system to supplement the needs of water consumption within the process. Similarly, Mussatto et al., (2013) in their techno-economic analysis also proposed the integration of reverse osmosis technology for treating wastewater in their biorefinery. The purified water was intended to partially substitute the water needs within the process, while the sludge was suggested to be used as a fertilizer. So, one possible strategy could be to integrate closed-loop wastewater recirculation after advanced water treatment into the current technology, which can substitute the cooling water requirements of the current process. This can simultaneously negate the environmental impacts of wastewater released into the environment along with cooling water use, thereby substantially enhancing the environmental sustainability of the process. This would further promote the circularity of the technology. However, the techno-economic feasibility, along with the associated environmental footprint of the wastewater treatment processes necessary to meet the cooling water quality standards needs to be stringently evaluated before implementation.

4.4 Scenario analysis

The present study observed the use of ultrapure water as cooling water utility to be one of the most significant input parameters as it has a massive contribution to the total environmental burden during LA production. The production of ultrapure water is immensely resource and energy intensive leading to a very high environmental footprint. So, the environmental sustainability of these LA production technologies could substantially improve with a shift towards other grades of water resources with relatively less purification. Hence, in this section, the base scenario of the ultrapure water usage was compared with two other scenarios using de-ionised (DI) water and completely softened water as

cooling water resource in different processes (Fig. 4a, 4b). Moreover, low pH fermentation with pinch technology which was determined as the most environmentally sustainable was selected for this scenario analysis for the purpose of ease. The DI water and completely softened water were selected based on their extensive use in industries as cooling water. It was observed that the electricity consumption for producing 1 kg of softwater and DI water are almost 100% and 88% lower than ultrapure water, respectively. Additionally, the water treatment processes used in the production of ultrapure water release considerably more contaminants into the environment compared to the other two grades of water.

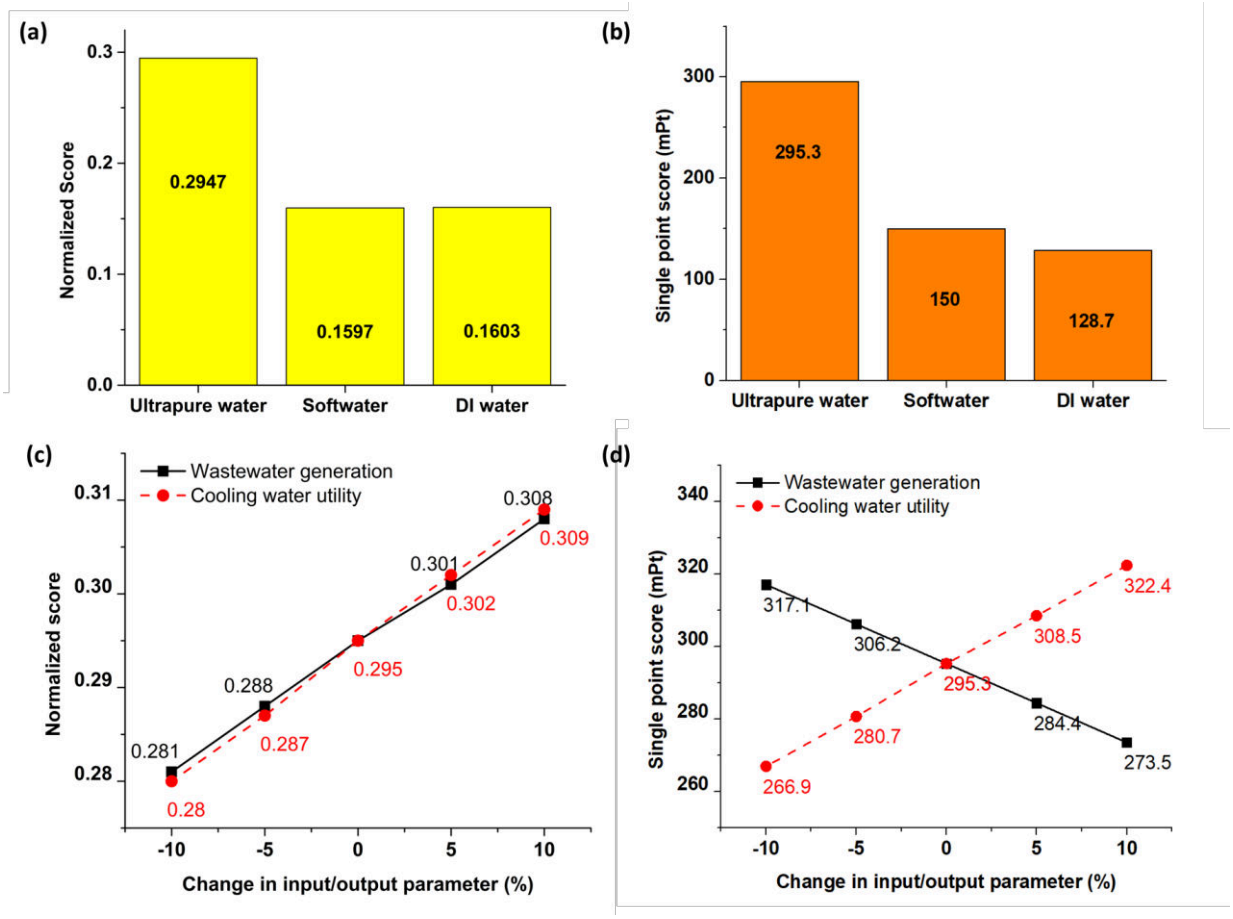


Fig. 4. Scenario analysis with respect to change in the type of cooling water used (a) midpoint impacts; (b) endpoint impacts; Sensitivity analysis with respect to change in the wastewater generation rate and cooling water utility (c) midpoint impacts; (d) endpoint impacts.

The use of DI water as an alternative to ultrapure water has reduced the midpoint and endpoint environmental impacts by 45.6% and 56.4%, respectively. Similarly, the use of soft water has resulted in a significant decrease in the midpoint and endpoint environmental impacts by 45.8%, 49.2%, respectively. Hence, a shift to a more sustainability sources of water for cooling purposes will positively influence the process and improve its overall environmental performance. However, the technical feasibilities associated with the shift need to be rigorously evaluated before application.

4.5 Sensitivity and uncertainty analysis

As evident from the mid and endpoint impact categories, wastewater generation from the AD process and cooling water utility emerged as the most significant output and input factors responsible for the environmental impacts in the process. So, sensitivity analysis was carried out for the low pH fermentation process by varying the wastewater generation from AD and cooling water utility by ± 5% and ± 10% (Fig. 4c, 4d). Low pH fermentation process with pinch technology was selected for sensitivity analysis as it was determined to be the most sustainable pathway. The sensitivity analysis has revealed that a reduction in the wastewater generation by 5% and 10% would reduce the overall midpoint environmental impacts of the process by 2.3% and 4.5%. In contrast, the endpoint impacts were increasing by 3.7% and 7.4% with decrease in wastewater generation by 5% and 10%, respectively. This negative correlation of the endpoint impacts could be attributed to the positive environmental significance of wastewater generation during AD in the midpoint impact category of water consumption.

Similarly, reducing the cooling water utility by 5% and 10% would decrease the midpoint impacts by 2.5% and 4.9%, and endpoint impacts by 4.9% and 9.7%, respectively. So, adopting more sustainable approaches, such as recycling the wastewater back into the system after treatment to compensate the water requirement for the entire process can be of positive influence in reducing the associated environmental burdens of water consumption and wastewater.

The uncertainty analysis for midpoint impacts of low pH fermentation with pinch technology was performed via Monte Carlo simulations. This was based on 10,000 simulation runs using a 95% confidence interval for the production of 1 kg LA from BW. The results depicted a relatively lower coefficient of variation in the major midpoint impact categories of concern, such as freshwater eutrophication (20.7%), marine eutrophication (21.6%), ozone formation on terrestrial ecosystems (16.5%), ozone formation on human health (16.5%), water consumption (-42.9 %), and global warming (12.8%).

5. Conclusions

This study analyses the relative environmental impacts of neutral and low pH fermentation techniques for the production of lactic acid (LA) from bread waste (BW). Anaerobic digestion (AD) and esterification were found to be the subprocesses responsible for the majority of the environmental effects. Especially, wastewater generation and cooling water utility were identified as critical environmental hotspots. Low pH fermentation technology was environmentally superior compared to neutral pH fermentation technology. Moreover, integration with pinch technology further enhanced the environmental performance of the process. Adopting sustainable practices for wastewater, and cooling water management such as integrating closed-loop wastewater recycling as a cooling water substitute within the process can further reduce the overall environmental footprint. This LCA can enable targeted environmental optimization of LA production and promote a sustainable and circular approach toward BW management. Given the increasing worldwide interest in the valorization of organic waste using biorefinery-based technologies, the environmental effects examined in our study could provide valuable insights into critical hotspots in similar processes used for generating various other value-added by-products. The findings of this study need to be further investigated and refined by developing sophisticated datasets for estimating the environmental footprint of the capital goods and infrastructural components. Moreover, comprehensive optimization of these major hotspot processes and impact categories to improve the environmental efficacy of biorefinery designs can be the focus of future

research. Furthermore, an LCA of an operational biorefinery in real-time could provide insights into the precision of the environmental footprint of a simulated counterpart.

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Author Contributions Statement

BKD, SKM and VK: Conceptualization, Supervision, Writing – review & editing; **KRV and RB:** Writing – original draft, Writing – review & editing; **SK:** Writing – review & editing.

KRV and RB carried out the experimental work, analyzed the data and wrote the Manuscript. BKD, SKM and VK provided useful suggestions for experimental design and revised the Manuscript critically. SK was involved in proofreading the Manuscript. All authors read and approved the final Manuscript.

Declaration of Interest Statement

The authors declare that they have no competing interests.

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Highlights

- Bread waste (BW), one of the major global food waste can serve as a sustainable feedstock for lactic acid (LA) production.
- Relative environmental performance of fermentative LA production technologies from BW was assessed using LCA.
- Low pH fermentation with pinch technology is more environmentally sustainable compared to neutral pH fermentation.
- Wastewater generation from anaerobic digestion and cooling water utility in esterification are environmental hotspots.
- This targeted environmental optimization study will aid policymakers in better decision-making in lactic acid production.

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Life cycle assessment of fermentative production of lactic acid from bread waste based on process modelling using pinch technology

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