Sustainable aviation fuel from Butanol: A Study in optimizing Economic and Environmental impact through process intensification

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ABSTRACT

Recent studies have provided a critical evaluation of the production of sustainable aviation fuels (SAFs) from biobutanol derived from biomass, underscoring their essential role in diminishing carbon emissions in the aviation industry. These studies highlight the significance of SAFs, particularly for long-haul flights, where they offer a viable solution for decarbonization and environmental sustainability. This process involves the conversion of biomass to butanol through the Acetone-Butanol-Ethanol (ABE) process and subsequent upgrading to SAF using the Alcohol-to-Jet (ATJ) pathway. Process intensification techniques have been integrated downstream with a simultaneous saccharification-fermentation (SSF) reactor, and upstream with a Petlyuk distillation column, aiming to optimize the system configuration to minimize Total Annualized Cost (TAC) and environmental impact as measured by the Eco-Indicator 99 (EI99) method. Metaheuristic optimization techniques were applied, leading to a process output of 91,358 kg/h of butanol and 45,833 kg/h of biojet fuel. The economic and environmental assessments of the process resulted in a TAC of USD 12,614/kg and an EI99 of $1.329x10^8$ points/kg. These findings suggest that the integrated biorefinery approach is a promising and efficient strategy for SAF production, contributing to the expansion of renewable energy sources in alignment with the United Nations' Sustainable Development Goals.

List of symbols

N_t	Total stages
N_f	Feed input stage
R_r	Reflux ratio
D_r	Distillate rate (kg/h)
SPF	Side product flow (kg/h)
SPF_{OS}	Side product output stage
LIF	Liquid interconnection flow (kg/h)
LIF_{IS}	Liquid interconnection flow input stage
LIF_{OS}	Liquid interconnection flow output stage
VIF	Vapor interconnection flow (kg/h)
VIF_{IS}	Vapor interconnection flow input stage
VIF_{OS}	Vapor interconnection flow output stage
V	Volume (m_3)
Q	Duty (MW)

W_F	Water flow (kg/h)
B_F	Biomass flow (kg/h)
EA_F	Extractant agent flow (kg/h)
E_F	Enzyme flow (kg/h)
H_F	Hydrogen flow (kg/h)
x_m	Liquid purity for the compound m
y_m	Vapor purity for the compound m
TAC	Total Annual Cost ($\$/kg$)
t_{ri}	Return-On-Investment Time (*years*)
C_{TM_i}	Total Cost of the i th process module ($\$$)
C_{ut_j}	Cost of the j-th service or input ($\$/year$)
F_k	Product flow k ($kg/year$)
n	Number of Process Units
m	Number of Services/Inputs
EI99	Ecoindicator 99 (*pts/kg*)
$\alpha_{b,k}$	Damage caused in category k for each unit of chemical B

released into the enviroment

β_b Total quantity of chemical B released per unit of reference flow

ω_d Weighting factor for the damage in each category d

δ_d normalization factor for the damage in category d

1. Introduction

Climate change emerges as a paramount challenge for humanity, predominantly propelled by the increasing emissions from fossil fuels [1]. Within this context, the aviation industry occupies a central role in the global economy, notwithstanding its substantial contributions to the anthropogenic greenhouse effect [2]. The advent of new technological solutions is imperative for the attainment of the sector's environmental objectives. The aviation industry is at a critical juncture in the global effort to address the escalating climate crisis, aligning its strategies with the United Nations' Sustainable Development Goals (SDGs), particularly Goals 7, 'Affordable and Clean Energy', and 12, 'Responsible Consumption and Production'. Set in 2015 with a target for achievement by 2030, these goals guide the industry's shift towards Sustainable Aviation Fuel (SAF) as an essential alternative to fossil-based jet fuels. Within the transportation sector, the aviation industry is identified as particularly challenging for the adoption of 'carbon neutrality' strategies. This complexity arises despite the fact that bioethanol, biobutanol and biodiesel have been introduced as substitutes for a portion of the gasoline and diesel fuel consumption, respectively [3,4]. In contrast, biofuels present a viable alternative for extended flight operations and have been integrated into the existing fleet as compatible fuels [5].

Currently, renewable jet fuel comprises a mere fraction, under 0.1 %, of the global demand for jet fuel, which was approximately 330 million tons in 2019 [6]. Efforts by certain governmental bodies and investment entities are underway to foster the production of low-carbon jet fuel, in light of aviation fuel's contribution to 3 % of worldwide carbon emissions [7,8]. Sustainable Aviation Fuel (SAF) has been internationally acknowledged as an effective strategy for diminishing the carbon and greenhouse gas emissions footprint of the aviation sector [9,10]. The process of converting commercially produced biobutanol into aviation-grade kerosene is at a critical juncture of development and opportunity [11,12].

Biobutanol involves the process of transforming biomass into fuel alcohol via microbial fermentation, wherein specific yeast species generate butanol as the principal product of fermentation [13]. As one of the products from acetone-butanol-ethanol (ABE) fermentation, Biobutanol also has the largest portion among all the solvents, Currently, the majority of fuel butanol is obtained through the fermentation process of food crops. This approach, however, is constrained in terms of scalability and sustainability in the long-term perspective. Hence, the emergence of second-generation biofuel butanol, derived from lignocellulosic biomass, stands as a critical factor for enabling the extensive substitution of petroleum-based fuels in future scenarios. Enhancing cost-effective and renewable biobutanol presents a viable alternative approach for the production of superior hydrocarbon fuels derived from renewable biomass sources [13].

Wang & Tao [12] have meticulously reviewed the array of process technologies pertinent to the production of SAF. Notably, conversion methodologies such as alcohol-to-jet (ATJ) [9,12], Fischer-Tropsch (FT) [10], hydrothermal liquefaction (HTL) [12], and hydroprocessed esters and fatty acids (HEFA) [5] are instrumental in transforming biomass into sustainable renewable jet fuel . These methodologies have either received certification or are currently under evaluation by the American Society for Testing and Materials (ASTM). A significant body of research, characterized as techno-economic analysis (TEA), has scrutinized the viability of these conversion technologies [12,14,15,16,17]. For instance, de Jong et al. [17] identified that the cost spectrum for SAF via technologies such as FT, ATJ, and HTL, tends to surpass that of traditional jet fuels. Additionally, numerous assessments, referred to as

life-cycle analysis (LCA), have been executed to calculate the greenhouse gas (GHG) emissions attributed to the deployment of these varied technological pathways [18,19]. Despite these thorough evaluations, it is significant to note that the integrated process viability of biobutanol, spanning from its inception as an alcohol to its conversion into SAF, remains insufficiently explored. Additionally, an analysis focusing on optimization criteria that evaluates sustainability indicators has not been undertaken, highlighting a critical gap in the current research landscape. This omission underscores the necessity for in-depth studies that not only assess the technical feasibility but also examine the sustainability impacts of biobutanol as a renewable energy source for aviation fuel.

The production of alcohols as biofuels from lignocellulosic material has seen a notable increase in developing countries in recent years, serving as a catalyst for advancing towards Sustainable Aviation Fuel (SAF) through the Alcohol-to-Jet (ATJ) process. Bioethanol, in particular, has demonstrated significant potential in reducing both the costs and emissions associated with the process. Nevertheless, the literature reveals a gap in the development of the ATJ process utilizing biobutanol, highlighting a strategic opportunity for the production of hydrocarbon fuels and by-products of interest such as acetone, ethanol, gasoline, and green diesel through this pathway.

Process intensification is a strategic approach to enhance production processes, offering cost and environmental impact reductions, along with improvements in operability and safety, product quality, waste reduction and process safety by applying innovative principle [13]. This is particularly relevant in liquid biofuel production, where processing renewable materials poses cost challenges. The goal is to boost biofuels' economic viability and achieve higher yields than conventional methods.Process intensification via upstream and downstream processing emerges as a critical strategy for augmenting the overall efficiency of continuous bioprocesses [20]. In the ABE process, downstream intensification can be achieved through the incorporation of simultaneous saccharification fermentation reactors, similarly, downstream enhancements can be implemented in the separation zone using Petlyuk column configurations to reduce both the costs and environmental impacts associated with the production of Sustainable Aviation Fuel (SAF) from biobutanol. These techniques have been successfully applied in HEFA and ATJ processes utilizing ethanol, resulting in significant reductions that steer these processes towards greater profitability. Therefore, applying such techniques to the ATJ process via biobutanol presents a viable and significant sustainable option.

Incorporating the Process Intensification (PI) philosophy into process optimization, the approach extends beyond traditional mathematical or metaheuristic optimization techniques, which focus on minimizing or maximizing an objective function within system constraints. By adopting PI, the strategy is not just about adjusting variables within the feasible region defined by process specifications and physical relations but about rethinking and integrating various operations and subsystems into a unified and optimized system. This holistic view aims to achieve greater efficiencies and innovations in industrial processes, leveraging the synergy between different parts of the process to enhance overall performance [21].The primary objective in sustainable aviation fuel production is to achieve an optimal design ensuring economic profitability while minimizing energy requirements and meeting strict sustainability criteria. An effective strategy to attain these goals involves the adoption of intensified zones, which reduce energy requirements, total annual cost, and environmental impact.

This study is distinguished by its thorough and detailed analysis of the sustainable aviation fuel (SAF) production process from biobutanol, leveraging advanced process intensification techniques to enhance sustainability. While ethanol has been the most extensively studied intermediate alcohol, n-butanol has emerged as a promising alternative. This methodology is further supported by a comprehensive and rigorous assessment of the process's sustainability, implementing strict sustainability criteria to establish a wholly sustainable methodology. Current

literature provides limited insights on optimizing SAF production from n-butanol in its standard configuration, particularly with sustainability criteria in mind. The novelty of this work lies in the application of process intensification techniques within an integrated biobutanol process, specifically within the context of a biorefinery. To date, no studies have been reported exploring potential process intensifications, nor have they been optimized towards sustainability objectives. By promoting the integration of these techniques at the reaction and purification stages, this study forges significant advancements in the efficiency and sustainability of the SAF production process, contributing to the pursuit of more effective and environmentally friendly renewable energy solutions.

2. General description

SAF production, especially when commencing with biomass as a foundational material, typically involves the transformation of biomass into intermediate alcohols. These processes, while innovative, are frequently challenged by high operational costs and significant energy consumption. Considering this, our research, referencing an analysis conducted in 2018, identifies sugarcane bagasse and corn straw as Mexico's most prolific biomass sources [22]. Leveraging data from the Agricultural and Fisheries Information Service and SAGARPA (2015), sugarcane bagasse emerged as the chosen raw material, mainly due to its abundant availability in Mexico during the year 2018. The calculation base used was 462,632.51 kg/h of sugarcane bagasse.

A pivotal aspect of this research is the development of a refined process tailored for the efficient extraction of vital sugars from sugarcane bagasse, crucial for the production of butanol. This process innovation is expected to reduce both the economic and energy footprints traditionally associated with SAF production. Table 1, an integral part of our study, outlines the detailed compositions of the considered biomasses, encompassing the percentages of cellulose, hemicellulose, lignin, ash, and moisture content [23]. The selection of sugarcane bagasse as the primary raw material not only aligns with the availability data but also serves as a model for utilizing locally abundant resources in SAF production.

The initial phase, biomass-to-butanol, incorporates innovative process intensification strategies, particularly focusing on the reaction and purification zones. These strategies are aimed at enhancing the efficiency and throughput of the process, while simultaneously minimizing the energy input and operational costs. In contrast, the subsequent butanol-to-biojet phase adheres to a more conventional processing framework. The process will be analysed in 2 subprocesses, the biomass-butanol process and the ATJ process.

A key aspect of this research is the application of an optimization methodology combined with process intensification techniques. This approach is anticipated to significantly diminish the total annual operational cost and curtail the environmental footprint of the production process. Furthermore, the study proposes a modular manufacturing paradigm. This modularity enables the strategic placement of the butanol production facility proximal to biomass harvesting sites, and the SAF production unit near airports. Such a configuration is not only cost-effective in terms of supply chain logistics but also offers enhanced flexibility, allowing the production network to adaptively respond to the fluctuating dynamics of supply and demand.

To elucidate the biomass-to-SAF conversion pathway, Fig. 1 is incorporated in this study. It graphically represents the integrated process flow, highlighting key stages, interconnections, and the transition from raw lignocellulosic biomass to the final biojet fuel product. This visual representation serves as a crucial tool for understanding the intricacies of the process and the potential impact of the proposed intensification and optimization strategies on the overall sustainability of biojet fuel production.

2.1. Biomass-butanol process

In the reaction zone, the process encompasses three crucial stages: pretreatment, enzymatic hydrolysis, and fermentation. The fermentation stage, critical for butanol production, is commonly referred to as ABE (Acetone-Butanol-Ethanol) fermentation. This process is notable for yielding acetone and ethanol as by-products. Contemporary research in butanol production has been increasingly concentrated on the integration of novel technologies aimed at augmenting the yield and energy efficiency of ABE fermentation, thereby enhancing the economic feasibility of butanol production. Clostridium acetobutylicum, a microorganism extensively studied and genetically modified for this purpose, plays a central role in the metabolic pathway. This pathway utilizes glucose and xylose as substrates, culminating in the production of the ABE mixture. A significant challenge in this process is the generation of solvent byproducts, which lead to cellular inhibition and eventually halt the production of the ABE mixture after a certain period [24].

The separation zone follows, wherein the fermentation broth is subjected to a series of purification steps. The objective is to isolate acetone, butanol, and ethanol with a target purity exceeding 99.5 % (w/w). However, this purification process encounters two major hurdles: the high dilution of the products in the fermentation broth and the presence of multiple azeotropes. Overcoming these challenges necessitates the application of advanced technologies such as gas stripping, pervaporation, and liquid-liquid extraction. These techniques are instrumental in breaking the azeotropes, thereby simplifying the subsequent separation process. Once the azeotropes are disrupted, the separation of the components becomes relatively straightforward, typically achievable through conventional distillation methods.

2.2. Butanol-to-SAF process

Dehydration is the initial step, primarily aimed at removing oxygen from the alcohol as water. In this process, C1 to C4 alcohols are transformed into alkenes, carbon compounds featuring double bonds, with lengths ranging from C2 to C5. This transformation is facilitated through a catalytic chemical reaction, often proceeding via the corresponding ether. The dehydration of tertiary alcohols is relatively straightforward, whereas primary alcohols necessitate higher temperatures, typically between 170 °C and 200 °C, in the presence of strong acids. The reaction temperatures for this dehydration process can span from 170 to 538 °C. This stage effectively removes the oxygen atom from the hydroxyl group – the defining functional group in alcohols – yielding alkenes suitable for further processing. The primary objective here is to optimize the production of short-chain alkenes while minimizing byproduct formation. For instance, the dehydration of n-butanol may result in the formation of cis- and trans-2-butene isomers.

The subsequent stage, oligomerization, involves the combination of short-chain molecules to form longer-chain hydrocarbons. In the context of the Alcohol-to-Jet (ATJ) process, C2 to C4 alkenes are converted into longer-chain alkenes with carbon numbers between 8 and 16. Several catalytic processes exist for ethene oligomerization, yielding linear chain alkenes. The choice of catalyst influences both the reaction conditions and the feedstock. Transition metal-based catalysts are particularly effective in liquid-phase oligomerization, while acid heterogeneous catalysts cater to other aggregation states and the production of branched alkenes with higher carbon atom counts [25].

Alkenes, characterized by their unsaturation and instability, are not
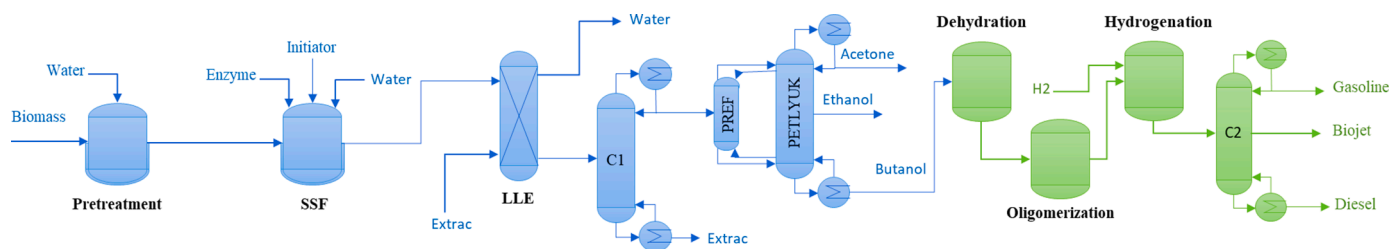
Table 1
Composition of biomass [23].

Biomass	Cellulose %	Hemicellulose %	Lignin %	Ashes %	Humidity %
Sugarcane bagasse	41	21	23	1.2	50
Corn stover	36	19	20	2.0	20

Fig. 1. Biomass-SAF process.

ideal for aviation fuel. Hence, hydrogenation is a critical step, aimed at converting alkenes into more stable alkanes by adding hydrogen. This process typically employs metal catalysts such as palladium, platinum, or nickel, dispersed on activated carbon. These catalysts are chosen for their high dissociation enthalpy, allowing the reaction to proceed under standard conditions. Raney-Nickel catalysts are commonly used in industrial settings. Depending on the catalyst used, the reaction temperature may need to be adjusted. Additionally, hydrogenation can be conducted at high pressures (200–350 bar), utilizing catalysts like copper-zinc chromite or sulfide, with temperatures ranging from 150 °C to 200 °C [25].

2.3. Optimization process

To conduct the global optimization, we employed a stochastic hybrid optimization technique known as Differential Evolution with Tabu List (DETL). Stochastic methods, in broad terms, have demonstrated their effectiveness in tackling complex optimization problems, which are often highly nonlinear and potentially non-convex, as evidenced by previous works [26,27]. DETL is rooted in the theory of natural selection. Initially proposed by Storn in 1997, Differential Evolution (DE) was originally designed for single-objective functions [28]. Subsequently, Madavan and Field (2002) adapted the method to address multi-objective problems [29]. The DE algorithm comprises four main steps: initialization, mutation, crossover, evaluation, and selection. During the initialization step, the algorithm begins its search in a d-dimensional space, starting randomly as follows:

$$\vec{X_{i,G}} = \left[X_{1,i,G}, X_{2,i,G}, X_{3,i,G}, \ldots, X_{D,i,G} \right] \tag{1}$$

Regarding the mutation step, which bears a striking resemblance to biological processes, mutation can be defined as a modification or perturbation with a random component. Beginning with a parent vector (referred to as the target vector), this parent vector undergoes mutation to produce a donor vector. Ultimately, the trial vector is derived by combining both the donor and target vectors. This process can be represented as follows:

$$\vec{V_{i, G}} = \vec{X_{r^1_{1, G}}} + F. \left(\vec{X_{r^1_{2, G}}} - \vec{X_{r^1_{3, G}}} \right) \tag{2}$$

Continuing to the crossover step, the target vector exchanges its components with the donor vector under this operation, resulting in the formation of the trial vector $\vec{U_{i, G}} = [u_{1, i\ G}, u_{2, i\ G}, u_{3, i\ G}, \ldots, u_{D, i\ G}]$. Thus, the trial vector is obtained as:

$$u_{j,i,G} = v_{j, i, G} \ for \ j = \ \langle n \rangle_D \ \langle n + 1 \rangle_D, \ \ldots, \ \langle n + L - 1 \rangle_D \tag{3}$$

$x_{j,i,G}$ for all other $j \in [1, D]$ Concerning the selection step, in order to maintain a constant population size, this step determines whether the target or the trial vector will persist from generation G to the subsequent generation $G + 1$. The selection process is outlined as follows:

$$\vec{X_{i, G+1}} = \vec{U_{i, G}} \ if \ f\left(\vec{U_{i, G}}\right) \leq f\left(\vec{X_{i, G}}\right) \tag{4}$$

$$\vec{X_{i, G+1}} = \vec{X_{i, G}} \ if \ f\left(\vec{U_{i, G}}\right) > f\left(\vec{X_{i, G}}\right) \tag{5}$$

Where $f(\vec{X})$ represents the objective function to be minimized or maximized.

Regarding Tabu concepts, both the Tabu list concept (TL) and Taboo Search (TS) as previously proposed by Glover in 1989, serve to prevent revisiting the search space by maintaining a record of visited points [30]. The TL is initialized randomly within an initial population and continuously updated with newly generated trial individuals. This taboo check occurs during the generation step of the trial vector, and the new trial individual is generated repeatedly until it is not in proximity to any individual in the TL. The total number of trial individuals, NP, is generated through the repetition of the steps. These newly generated NP trial vectors are then combined with the parent population to create a combined population totalling 2NP individuals.

The Differential Evolution with Tabu List (DETL) method offers numerous advantages, particularly in the realm of Global Optimization. DETL excels in conducting global optimization, enabling it to explore the entire search space for the optimal solution rather than being confined to local optima. Its stochastic nature lends robustness and versatility, providing a significant edge in addressing complex, highly nonlinear, and potentially non-convex optimization challenges. By combining the exploratory capabilities of Differential Evolution (DE) with the exploitative strengths of the Tabu List (TL), DETL adeptly navigates the search space while avoiding redundant exploration of previously visited solutions. Furthermore, DETL offers adaptability through adjustable parameters such as population size, maximum number of generations, crossover probability, and mutation factor, providing a distinct advantage by catering to diverse problem domains and facilitating optimization flexibility. Additionally, this hybrid platform seamlessly integrates with Aspen Plus and Microsoft Excel, offering users the benefit of a user-friendly interface and enabling effortless implementation and incorporation into existing simulation and optimization workflows.

However, the Differential Evolution with Tabu List (DETL) method also presents several disadvantages. First, DETL is sensitive to parameter selection, including factors such as population size, crossover probability, and mutation factor, necessitating meticulous adjustment for optimal performance and consuming considerable time and computational resources. Second, DETL exhibits high computational complexity, particularly noticeable in scenarios with extensive search spaces or intricate objective functions, resulting in longer optimization durations and increased computational demands. Third, the substantial memory resources needed to maintain the Tabu List (TL), especially noticeable in situations involving large populations or prolonged optimization runs, constitute a significant drawback, potentially limiting DETL's scalability for addressing exceedingly large-scale optimization problems. Fourth, despite its purported global optimization capabilities, DETL may still converge to local optima, especially evident in rugged search spaces or those containing multiple local optima near the global optimum, representing a significant disadvantage. Finally, DETL's effectiveness depends on the specific characteristics of the optimization problem domain, limiting its applicability, as its efficacy can vary, making it suboptimal for certain scenarios.

3. Methodology

The methodology for producing sustainable aviation fuel (SAF) is outlined below, divided into two main parts: biomass-butanol and butanol-SAF (ATJ process). Subsequently, the optimization approach employing The Differential Evolution with Tabu List (DETL) method is also elucidated.

3.1. Biomass-butanol process

The modeling of the butanol production process encompasses various components. The pretreatment reactor was modeled based on the yields, incorporating data from Table 2. The SSF reactor, a key element of the process, was modeled using kinetic models for enzymatic hydrolysis and fermentation. These models were simultaneously programmed in MATLAB software, integrating the complex dynamics of the biochemical reactions. The purification stage, in contrast, was modeled using Aspen Plus 8.8, a software renowned for its capabilities in chemical process simulation. The integration of MATLAB and Aspen Plus was achieved through the use of macros in Visual Basic, ensuring seamless data transfer and process simulation.

The kinetic models for enzymatic hydrolysis and fermentation, fundamental to the effective operation of the SSF reactor, are elaborated in this study. Additionally, material balances and associated parameters crucial for the process design are detailed in supplementary material A. This comprehensive approach to modeling the butanol production process not only exemplifies the application of advanced computational tools but also highlights the importance of process integration and intensification in the pursuit of economically viable and environmentally sustainable aviation fuel production.

For the purification stage, the focus shifts to the implementation of an intensified distillation system. Thermally coupled distillation systems are considered for their potential to yield significant economic and energy savings. These systems, characterized by their enhanced heat integration and reduced energy requirements, are pivotal in streamlining the separation process.

For the conversion of butanol from lignocellulosic biomass, the selection of an appropriate pretreatment method is crucial. This study, echoing the scientific rigor and analytical approach, examines two predominant pretreatment techniques: Liquid Hot Water (LHW) treatment and alkaline treatment, particularly focusing on their applicability to sugarcane bagasse (Fig. 2).

Liquid Hot Water Pretreatment (LHW) leverages elevated temperatures and pressure to maintain water in a liquid state, effectively fractionating the biomass. The efficacy of LHW is evidenced by its ability to produce a highly reactive cellulose fiber. This method facilitates the recovery of most pentoses while limiting the generation of degradation products that could potentially impede subsequent hydrolysis and fermentation processes [31]. On the other hand, alkaline pretreatments, typically employing sodium hydroxide, enhance the digestibility of cellulose and are more effective in solubilizing lignin [32]. These treatments result in a swelling of the biomass, thereby increasing the internal surface area of cellulose and decreasing its degree of polymerization and crystallinity. Such alterations in the lignin structure

contribute to the overall efficiency of the pretreatment process [33].

A comprehensive analysis of various pretreatment methods was conducted, with particular attention to their yields when applied to sugarcane bagasse (BCA). Table 2 in the study delineates the outcomes of each pretreatment method, including the residual solids and their respective glucan, xylan, and lignin contents [34]. The criteria for selecting an optimal pretreatment method were based on several key factors: high glucan conversion rates, efficiency in alcohol production, and the performance ratio in intensified systems like Simultaneous Saccharification and Fermentation (SSF) [35]. Among the evaluated pretreatment methods, Liquid Hot Water (LHW) emerged as an efficient and economically viable option for the selected raw material, sugarcane bagasse.

During the pretreatment stage, key biochemical reactions occur, primarily involving the breakdown of complex carbohydrates present in the biomass. The primary reactions modeled in this process are:

$$Glucan + H_2O \rightarrow Glucose \tag{6}$$

$$Xylan + H_2O \rightarrow Xylose \tag{7}$$

These reactions are fundamental in converting the polysaccharides present in the lignocellulosic biomass into fermentable sugars. The simulation of these reactions was conducted using a performance reactor model. This approach allows for the assessment of the efficiency and effectiveness of the pretreatment process under defined operational conditions.

The data utilized for modeling the LHW pretreatment process were sourced from Table 2, providing empirical insights into the yields and efficiencies of the pretreatment under various conditions. The LHW pretreatment, as modeled in this study, is executed at a temperature of 180 °C [34]. This temperature is selected based on its effectiveness in facilitating the hydrolysis reactions while minimizing the formation of inhibitory by-products that could adversely affect downstream processes like fermentation.

This modeling approach not only helps in understanding the chemical transformations occurring during the pretreatment but also aids in optimizing the conditions for maximum yield of fermentable sugars. The accuracy and effectiveness of this pretreatment modeling are crucial for ensuring the overall efficiency and sustainability of the butanol-to-SAF production process.

The study proposes the exploration of a Simultaneous Saccharification and Fermentation (SSF) reactor. The implementation of SSF aims to amalgamate enzymatic hydrolysis and fermentation into a single step, potentially leading to reductions in both energy consumption and economic costs compared to conducting these operations separately. The SSF process represents a strategic integration of biochemical reactions, enhancing overall process efficiency.

The kinetic model for cellulose hydrolysis is designed to predict the process performance under a variety of operating conditions efficiently. This hydrolysis involves a sequence of three reactions, each leading to the production of glucose. A notable aspect of these reactions is the potential for inhibition by the glucose produced or by other sugars already present in the system [36]. Such inhibition phenomena play a critical role in the kinetics of enzymatic hydrolysis and must be carefully

Table 2
Compositions of pretreated SCB [34].

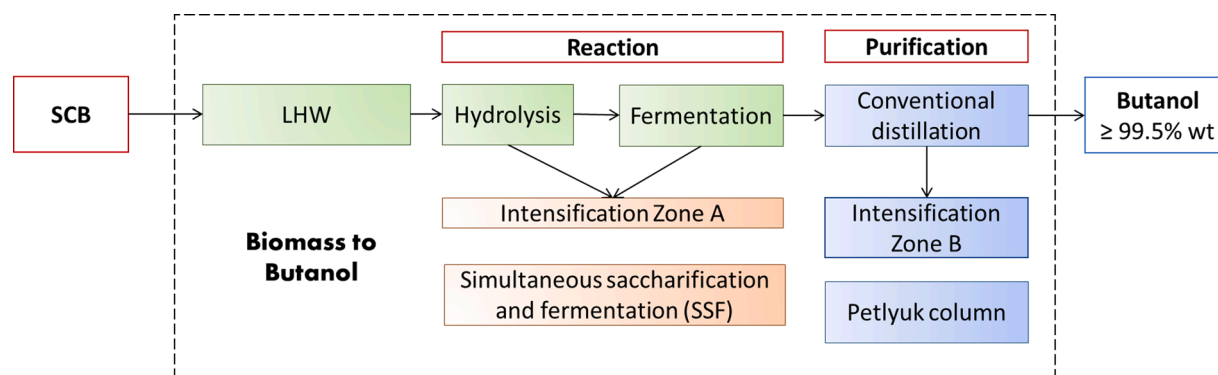| Pretreatment	Solid remain%	Glucan		Xylan		Lignin	
		Content%	Recovery%	Content%	Recovery%	Content%	Recovery%
LHW	59.05	63.12	88.44	6.49	82.35	23.07	42.31
NaOH	61.20	63.19	92.18	26.63	24.88	8.55	77.85
LHW + NaOH	42.5	82.45	83.53	3.97	92.22	13.07	76.47
NaOH + LHW	45.55	76.91	83.50	13.75	71.15	8.13	84.33

Fig. 2. Intensification Zones.

considered in the model.

For the purpose of this study, the enzymatic hydrolysis was modeled under specific operating conditions. These conditions include a temperature setting of 50 °C and a residence time of 72 h [36]. The choice of these parameters is based on their effectiveness in facilitating the enzymatic breakdown of cellulose, while also balancing factors like enzyme stability and reaction rates. Fig. 3 in the study presents a detailed kinetic model of the enzymatic hydrolysis process [36]. This model incorporates the enzymatic reaction mechanisms, taking into account the potential inhibitory effects of the sugars. It provides a comprehensive understanding of the dynamics of cellulose breakdown, which is essential for optimizing the hydrolysis process in terms of efficiency and yield, Additional equations and detailed information can be found in the supplementary material A.

The metabolic pathway of Clostridium acetobutylicum, as employed in this study, is based on the model proposed by Shinto in 2008. This comprehensive model considers both xylulose and glucose as substrates and encompasses a total of 17 distinct biochemical reactions. One of the significant strengths of this model is its ability to predict the production levels of acetone, butanol, and ethanol, which are critical outputs of the ABE fermentation process [37].

For the purpose of this study, the fermentation process was simulated under specific operating conditions. These conditions include maintaining a temperature of 35 °C and a residence time of 104 h [24]. These parameters were carefully chosen to optimize the growth and metabolic activity of Clostridium acetobutylicum, thereby enhancing the efficiency and yield of the ABE fermentation process. Fig. 4 in the study illustrates the kinetic model of Clostridium acetobutylicum [24]. This model is integral to understanding the metabolic pathways and reaction kinetics within the fermentation process. It provides detailed insights into how the substrates are converted into the desired products, and how various

factors like temperature and substrate concentration can impact the efficiency of the process.

The conceptualization of the SSF reactor is grounded in the principle of unifying the kinetic models of hydrolysis and fermentation. This integration aims to simulate both saccharification and fermentation within a singular operational unit. The development of this novel kinetic model for SSF is undertaken using MATLAB, employing the fourth-order Runge-Kutta method for its computational solution. This approach allows for a precise and robust simulation of the SSF process, taking into account the complex interplay of enzymatic breakdown and microbial fermentation. The implementation of an intensified SSF reactor is anticipated to bring about a host of benefits. These include a reduction in the number of required processing units, leading to decreased overall production costs. Additionally, such a system is expected to enhance raw material utilization efficiency, reduce the consumption of utilities, and achieve higher yields, all while realizing significant energy savings. This streamlined approach contrasts sharply with conventional configurations, which typically involve separate units for pretreatment, enzymatic hydrolysis, and fermentation, The complete set of equations and additional information can be found in the Supplementary Material A.

To further optimize the reaction zone, the pretreatment reactor is incorporated into the intensified SSF system. In contrast to the conventional tripartite setup, the intensified configuration consolidates the process into two reactors: one for pretreatment and the other for the combined saccharification and fermentation. This reduction in reactor count not only simplifies the process but also enhances its overall efficiency and sustainability.

Fig. 5 in the study illustrates the reaction scheme of this intensified process. This schematic representation provides a clear and concise visualization of the integrated SSF system, highlighting the streamlined pathway from biomass pretreatment to the production of fermentable sugars and their subsequent conversion into biofuel intermediates. The modeling and implementation of this intensified SSF reactor embody a significant stride towards optimizing the butanol-to-SAF production process, showcasing a sophisticated integration of biochemical engineering and process intensification strategies.

Unconventional distillation techniques, particularly those employing thermally coupled systems, offer significant energy savings and process efficiency. These systems utilize thermal coupling, where heat transfer occurs through direct contact between material flows within the columns. In the context of ternary mixtures like ABE, thermally coupled arrangements such as the side stripper (TCDS-SS), side rectifier (TCDS-SR), and the fully thermally coupled Petlyuk scheme are considered. Comparative studies of these systems, demonstrate potential energy savings of up to 30 percent compared to conventional distillation methods. The Petlyuk system (Fig. 6) exhibits superior energy efficiency compared to systems with side columns [38].

The design of the Petlyuk distillation system begins with a reference to the conventional distillation setup. Both systems are initially designed
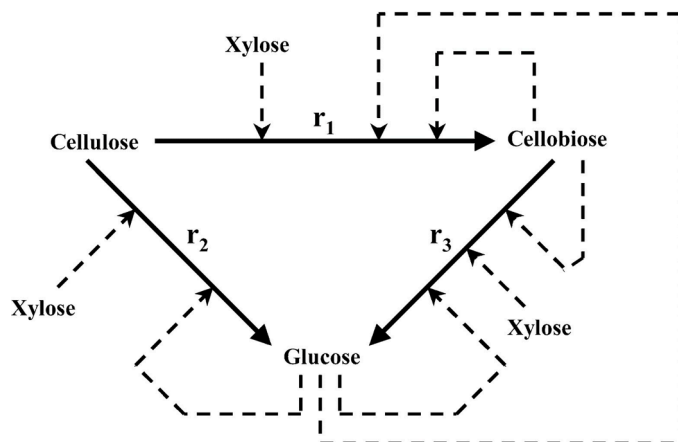

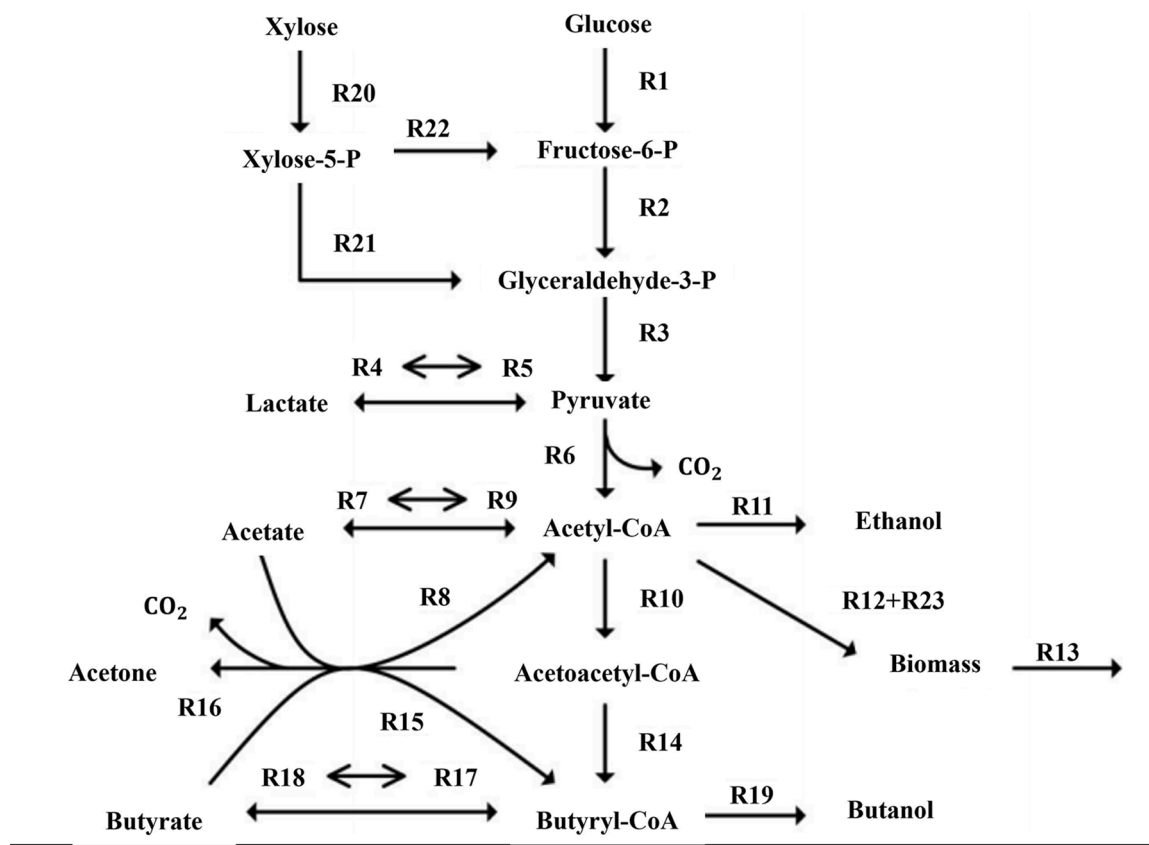**Fig. 3.** Kinetic model of the enzymatic hydrolysis [36].

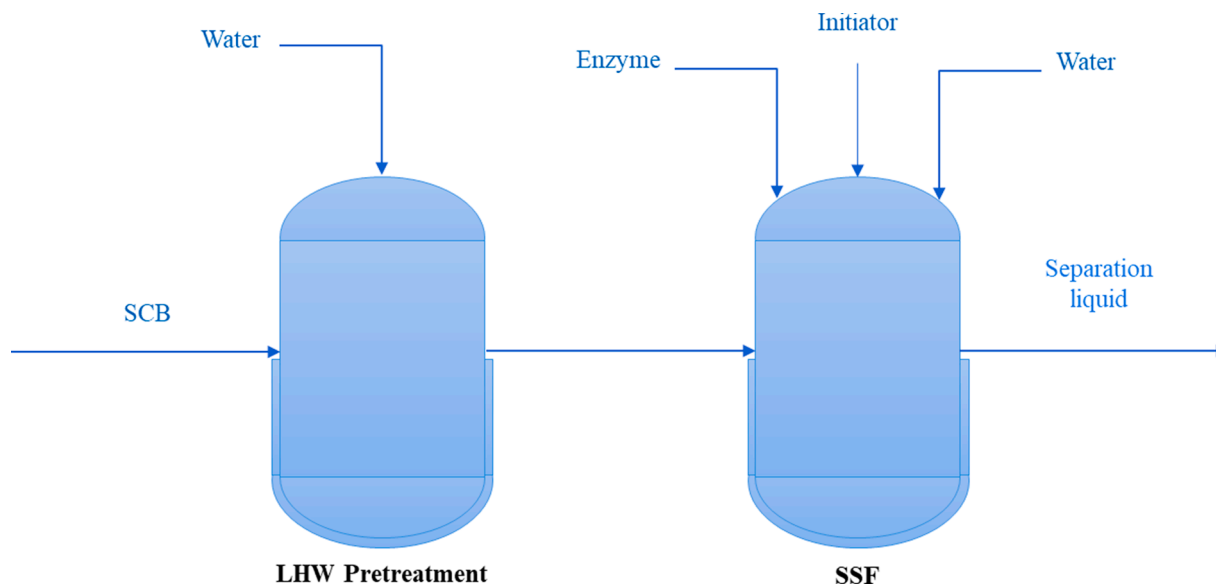**Fig. 4.** Kinetic model of clostridium acetobutylicum [24].

Fig. 5. Reaction scheme.

using the Aspen Plus simulator. Preliminary designs are created using shortcut methods (DSTWU module), followed by more rigorous methods (RADFRAC module). The Non-Random Two-Liquid (NRTL) thermodynamic model is selected due to the presence of azeotropes in the ABE mixture. Fig. 7 illustrates the experimental composition of the n-butanol/water binary system obtained within the temperature range of 323−393 K and pressures ranging from 13.4 to 267 kPa [30], alongside data predicted by the NRTL model using default Aspen Plus binary interaction parameters. Additionally, prior studies have noted a

relatively high accuracy in predicting interactions between the components [39,40,41,42]. Rigorous simulations involve using a Kettle reboiler and total condenser. Key parameters such as the total number of stages, feed stages, reflux ratio, and column diameters are optimized. The design process involves rearranging sections of the conventional system to derive a preliminary design for the Petlyuk system, subsequently leading to a detailed design process. The ultimate aim of this intensification is to align with sustainability goals, reducing energy requirements and lowering the total annual cost of butanol production.
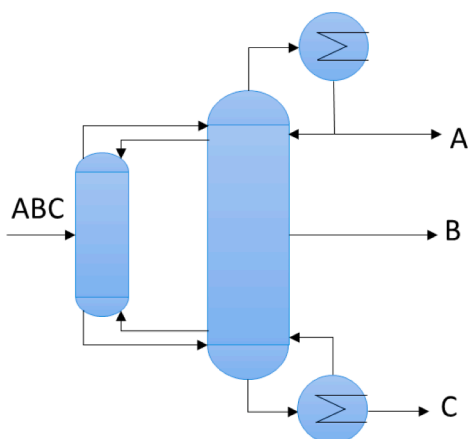
Fig. 6. Fully thermally coupled system (Petlyuk system).

To address the challenge of breaking azeotropes in the ABE mixture, a liquid-liquid extraction (LLE) column is introduced, utilizing hexyl acetate as the extracting agent. Additionally, a distillation column is integrated for the recovery and reuse of the extracting agent. The complete purification system, encompassing both the LLE and distillation components, is depicted in Fig. 8.

3.2. Butanol-to-SAF process

The Alcohol-to-Jet (ATJ) process, pivotal in converting butanol to Sustainable Aviation Fuel (SAF), involves a series of complex chemical transformations. This study employs Aspen Plus software, integrated with the Non-Random Two-Liquid (NRTL) thermodynamic model, to simulate these transformations. The NRTL model is particularly adept at handling non-ideal mixtures common in biofuel production processes.

Catalysts play a pivotal role in the conversion of alcohols into hydrocarbons, a crucial aspect of the research. The exploration of dehydration catalysis commenced with the utilization of materials like alumina and transition metal oxides but evolved to incorporate silicoaluminophosphates (SAPO), H-ZSM-5 zeolite catalysts, and heteropolyacid catalysts in subsequent studies [16]. Among these catalysts, the

0.5 %La–2 %P H-ZSM-5 catalyst has emerged as the most promising candidate, demonstrating exceptional results such as nearly complete conversion and an impressive 99.9 % selectivity toward ethylene when operated at 250 °C with a weight hourly space velocity of 2 h^{-1} [14,15]. The dehydrated ethylene produced can undergo further processing through a catalytic oligomerization technique to yield valuable linear α-olefins [18]. In the dehydration stage, butanol undergoes a critical reaction to form butene [19]:

$$C_4H_9OH \rightarrow C_4H_8 + H_2O \tag{8}$$

Simulated as a yield reactor, this stage operates at 35 °C and 1 atm. The primary goal is to remove water, thus enhancing the feedstock's suitability for subsequent reactions. This stage's efficiency critically influences the overall yield and quality of the final SAF product.

Contemporary industrial methods for synthesizing α-olefins encompass a spectrum of both homogeneous and heterogeneous catalysts. Diverse catalysts, including Ziegler Natta-type catalysts, chromium diphosphine catalysts and zeolites, have undergone extensive research and investigation [44]. For instance, employing a Ziegler Natta-type catalyst at temperatures ranging from 90 to 110 °C and pressures of 89 bar, a remarkable 96–97 % yield of linear α-olefins within the C4–C20 carbon range was achieved [45]. In industrial-scale oligomerization procedures, broader carbon number distributions are generated, yielding approximately 5 % C4; 50 % C6–C10; 30 % C12 and C14; 12 % C16 and C18; along with 3 % C20 and C20þ hydrocarbons [44], operating at elevated temperatures of 200 °C and pressures of 250 bar. The resultant olefin products are subsequently subjected to distillation processes to obtain diesel- and jet-range fuels, as well as light olefins [46]. This catalytic system facilitates the oligomerization of butene, a process essential for creating longer hydrocarbon chains. The reaction can be represented as:

$$m[C_4H_8] \rightarrow C_{4m}H_{8m} \tag{9}$$

where "m" varies from 2 to 4. The process yields a mixture of hydrocarbons, crucial for the desired fuel properties of SAF. The approximate distribution of the products is 36 % diisobutene, 22 % C8 dimers, 38 % triisobutene, and 4 % tetraisobutene [46].

Hydrogenation is a fundamental step in the production of SAF. It involves the addition of hydrogen to unsaturated hydrocarbons, thereby
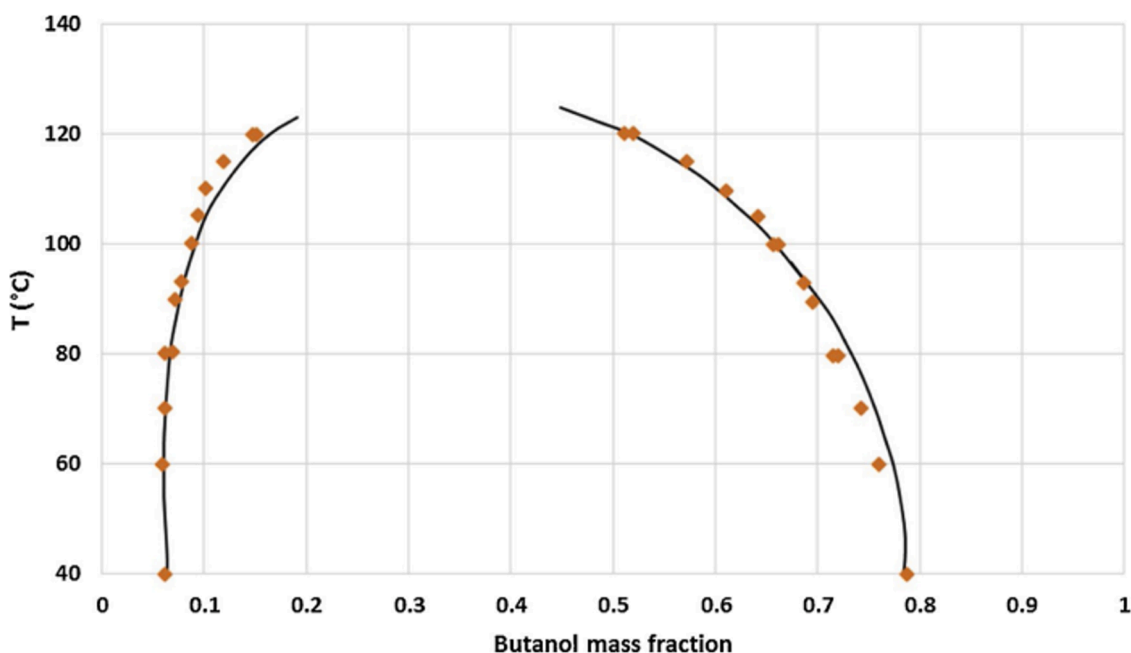

Fig. 7. Comparison between LLE predicted by NRTL model (line) and experimental data [43].
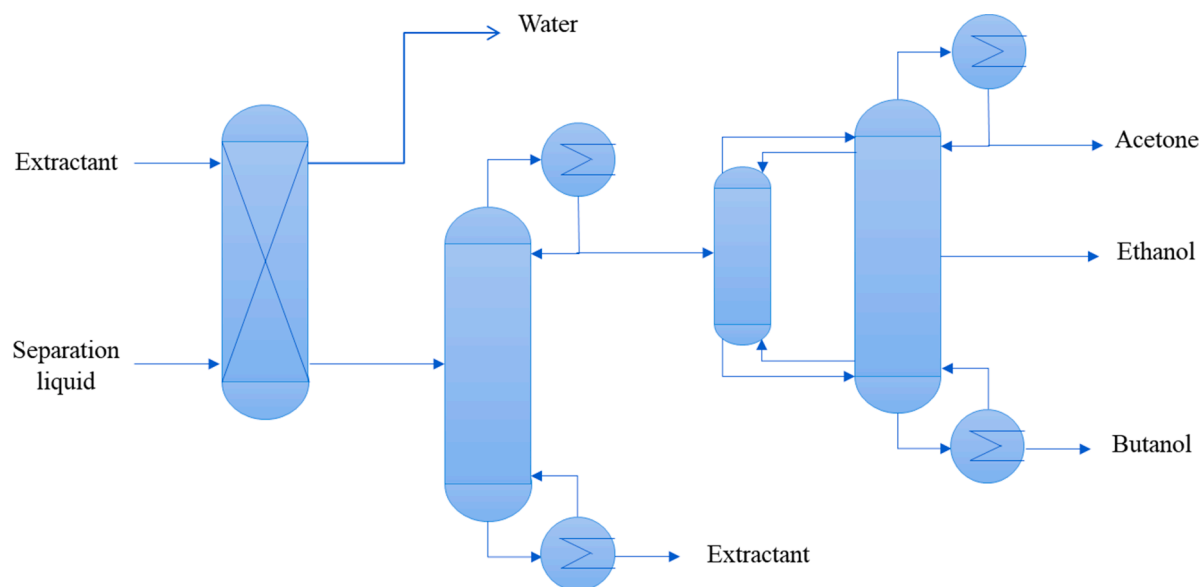
Fig. 8. Separation scheme biomass-butanol process.

increasing their saturation and improving fuel stability and quality. The reactor conditions and catalyst selection for this stage are crucial for achieving the desired product specifications. To produce renewable jet fuels, products within the jet fuel range (C9–C16) can undergo a hydrogenation process. This hydrogenation process is carried out at elevated temperatures of 370 °C and a Weight Hourly Space Velocity (WHSV) of 3 h1, with hydrogen supplied in amounts exceeding 5 % by weight. The catalyst employed for this process consists of either palladium or platinum supported on activated carbon [45]. As a result of the hydrogenation step, the C9–C16 alkanes generated are well-suited for the production of sustainable jet fuels.

The distillation column, designed using rigorous methods (RAD-FRAC) and based on the NRTL model, separates the final products: gasoline, biojet, and diesel. The column configuration enables the efficient separation of these components, with gasoline as the distillate, diesel in the bottoms, and SAF as a side product. The effectiveness of this separation process is vital for the economic viability and quality of the produced fuels.

3.3. Process optimization

In the framework of a circular economy, the conversion of lignocellulosic biomass into biojet fuel emerges as a complex optimization problem, necessitating the meticulous selection of appropriate technologies and the fine-tuning of design and operational parameters. This optimization is dual-pronged, aiming to maximize economic efficiency while simultaneously minimizing environmental impacts. The achievement of these objectives is pivotal in establishing a process that is both economically viable and ecologically sustainable. Furthermore, a process design characterized by minimal energy requirements, coupled with maximized economic savings, aligns strategically with the Sustainable Development Goals of the United Nations' 2030 Agenda, specifically targeting Goals 7 (Affordable and Clean Energy) and 12 (Responsible Consumption and Production). For evaluating the sustainability of the biojet fuel production process, this study employs two primary indicators: Total Annual Cost (TAC) and Ecoindicator 99 (EI99),

representing economic and environmental dimensions, respectively.

The TAC is a comprehensive economic metric reflecting the aggregate annual operational and capital expenditures of the biojet production process. As an indicator, it encompasses not only the product-related costs but also the broader process characteristics, offering a more informative comparison. The formula for calculating TAC, as outlined in Eq. (10), integrates the costs of heating, cooling, electricity, and inputs, alongside the capital costs of equipment [47,48,49].

$$TAC(USD/kg) = \frac{\frac{\sum_{i=1}^{n} C_{TM_i}}{t_{ri}} + \sum_{j=1}^{M} C_{ut_j}}{F_k} \tag{10}$$

This comprehensive equation thus encapsulates the total cost incurred in producing a unit weight of the product, considering both capital investment and operational expenditures. The prices of the supplies are provided in additional equations and detailed information can be found in the supplementary material B.

The EI99, an integral part of the life cycle analysis methodology, quantitatively assesses the environmental impact of a product or process. It encompasses human health, ecosystem quality, and resource depletion. The EI99 calculation, as per Eq. (11), aggregates the ecological impacts of chemical emissions across these categories [49,50].

$$EI99(ecopts/kg) = \frac{\sum_b \sum_d \sum_{k \in K} \delta_d \omega_d \beta_b \alpha_{b,k}}{F_k} \tag{11}$$

The EI99 value, thus, provides a holistic measure of the environmental burden of a process or product, considering a range of ecological impacts. EI99 is measured in ecopoints, with one ecopoint equating to one-thousandth of the average annual environmental burden of a European citizen. supplementary material C details the impact categories and their unit values.

The objective functions for assessing the sustainability of the process are framed in terms of minimizing the TAC and EI99. To align with the SDGs 7 and 12, the process aims to reduce both energy requirements and operational costs while concurrently lowering the environmental impact. The objective functions are formulated as:

$$Min(TAC_{tot}, EI99_{tot}) = f(N_t, N_f, R_r, LIF_{IS}, VIF_{IS}, LIF_{OS}, VIF_{OS}, LIF, VIF, SPF_{OS}, SPF, D_r) \tag{12}$$

Subject to $\overrightarrow{y}_m \geq \overrightarrow{x}_m$

In this study, we introduced a novel hybrid multi-objective optimization approach for producing Sustainable Aviation Fuel (SAF). This innovative approach amalgamates the principles of Differential Evolution (DE) with tabu meta-heuristics, resulting in an advanced evolutionary optimization technique. A key innovation in this method is the incorporation of a taboo list (TL) into DE, as conceptualized by Srinivas and Rangaiah. This integration prevents the algorithm from re-evaluating previously assessed regions of the search space, leading to the development of a more efficient hybrid stochastic optimization method termed DETL. The inclusion of the TL significantly improves the Differential Evolution algorithm's efficiency by eliminating redundant evaluations, which in turn reduces the computational time required for global optimization.

Sharma and Rangaiah [51], expanded upon this framework to tailor it for multi-objective optimization challenges, achieving promising outcomes. The resulting MODE-TL algorithm, a fusion of multi-objective DE and TL, has been proven effective by Sharma and Rangaiah in addressing intricate multi-modal optimization issues. This multi-objective optimization was carried out using a hybrid platform that integrates a Microsoft Excel implementation of MODE-TL with Aspen Plus, a method that has been previously established and validated by several researchers. In implementing the MODE-TL method, parameters included 200 individuals, 500 generations, a taboo list that accounts for 50 % of the population, a taboo radius of 2.5^{-06}, and crossover and mutation rates of 0.8 and 0.6, respectively. These parameters were optimized through an initial tuning process involving preliminary calculations.

This optimization algorithm's deployment utilizes a hybrid platform comprising Microsoft Excel for algorithm programming, a link to Matlab for bioreactor modeling, and a connection to Aspen Plus for simulating the separation unit. Decision variable vectors, or design variables, are transmitted from Microsoft Excel to Matlab using Dynamic Data Exchange (DDE), facilitating bioreactor simulation and subsequent input flow provision to the separation unit. Post-simulation, Matlab and Aspen Plus feed the resultant vector back to Microsoft Excel, which then analyzes objective function values to propose new decision variable values

per the utilized stochastic optimization method (Fig. 9). The DETL method employed parameters such as 120 individuals, 500 generations, a Tabu list covering 50 % of the total individuals, a Tabu radius of $1x10^{-8}$, and crossover and mutation fractions of 0.85 and 0.5, respectively, fine-tuned through a process of initial calculations. Objective functions were subjected to a range of values and decision variable restrictions outlined in Table 3, established pre-optimization to ascertain each variable's ranges.

Generally, within a multi-objective optimization framework, determining the optimal sequence that aligns with sustainability and green chemistry principles can pose challenges. Thus, this study endeavors to offer a more comprehensive perspective on design selection through the implementation of a normalization procedure. Normalizing the objective functions enables the identification of sequences that best adhere to sustainability principles. Koski (1985) proposes the following normalization approach for the objective functions [52]:

$$f_i(x) = \frac{F_i(x) - \min F_i(x)}{\max F_i(x) - \min F_i(x)} \quad i = objective\ functions,\ X \in NP \quad (13)$$

When considering points as vectors originating from the origin in a plane or space, it becomes feasible to compute the length of each vector to identify the one that minimizes both axes. By applying the Pythagorean Theorem, the Euclidean distance can be calculated universally as follows:

$$Distance = \sqrt{\sum_{i=1}^{i=NF} f_i^2},\ NF = Number\ of\ objective\ functions \quad (14)$$

Employing this approach of global selection criteria transforms the multiobjective optimization problem into the optimization of a scalar to determine the optimal solution. The advantage of normalization lies in its ability to indicate the distance between the optimum of an objective function for one sequence and the optimum of the same objective function for another sequence. In essence, it illustrates the extent to which we deviate from the ideals of a sustainable process.
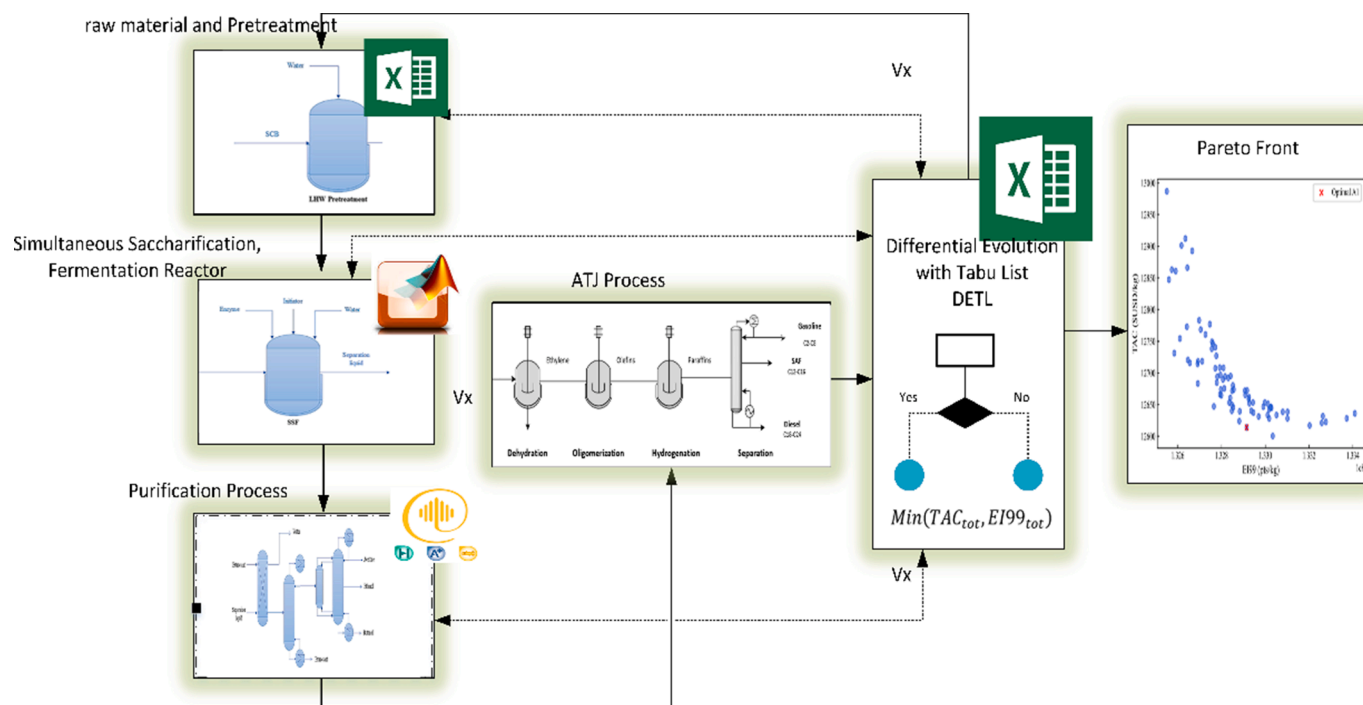

Fig. 9. Optimization process flow chart.

Table 3

Decision variables.

Variable	Range					Type
	LLE	*C1*	*Prefractionator*	*Petlyuk*	*C2*	
N_t	5–10	35–45	8–15	55–65	15–45	*Discrete*
N_f		15–30	3–7		2–40	*Discrete*
R_r		0.9–1		4–7	195–205	*Continuous*
LIF_{IS}				40–50		*Discrete*
VIF_{IS}				10–20		*Discrete*
LIF_{OS}				10–20		*Discrete*
VIF_{OS}				40–50		*Discrete*
SPF_{OS}				25–35	2–30	*Discrete*
LIF				22,666.66–23,859.64		*Continuous*
VIF				48,095.23–50,626.56		*Continuous*
SPF				13,454.45- 14,127.17	43,391.06–45,674.81	*Continuous*
D_r		138,638.78–145,935.56		38,373.79–40,393.46	18,836.80–20,929.78	*Continuous*

4. Results

The application of a differential evolution method with a tabu list for multi-objective stochastic optimization has yielded significant results in the production of butanol for sustainable aviation fuel. Stringent purity constraints were imposed on the biomass-butanol process, requiring at least 99.5 % purity for key components such as acetone, butanol, ethanol, water, and the extractant agent. In the Alcohol-to-Jet (ATJ) process, the distillation column's operational constraints were tailored to produce specific hydrocarbon chain lengths: short-chain hydrocarbons (C_8,) at the dome, medium-chain hydrocarbons (C_{12} to C_{16}) as the side product, and longer-chain hydrocarbons ($C_{20} - C_{32}$).at the bottom.

The optimized design for the biomass-butanol process, detailed in Table 4, integrates Liquid Hot Water (LHW) pretreatment, an intensified Simultaneous Saccharification and Fermentation (SSF) reactor, and a Petlyuk column for separation. A notable aspect of this design is the significant energy requirement of the reboiler duty in column C1, tasked with recovering the extractant agent. This optimized process design is capable of producing 91,358 kg/h of butanol, primarily from the bottom stream of the Petlyuk column.

In the conventional butanol-biojet (ATJ) process, as shown in Table 5, the design facilitates the production of 45,833 kg/h of biojet fuel, which is extracted as the side product of column C2, utilizing the butanol produced in the preceding process.

Fig. 10 displays the results obtained by the optimization method; the figure represents the Pareto front corresponding to the evaluated objective functions. From 81,000 evaluations (120 individuals and 200 generations) onward, no substantial improvements were observed,

Table 5

Design parameters for butanol-biojet processes.

Parameter	Equipment			
	Dehydration	*Oligomerization*	*Hydrogenation*	*C2*
N_t	–	–	–	17
N_f	–	–	–	15
R_r	–	–	–	195.32
SPF_{OS}	–	–	–	8
SPF	–	–	–	45,615.18
D_r	–	–	–	20,721.48
V	3515.46	3369.95	13.3339894	–
Q	29.95	–	14.59	351
H_F	–	–	555.02	–

leading to the assumption that the optimal zone for minimizing the evaluated objective functions (TAC and EI99) has been identified. The determined optimal point A1 reported values of 12,614 USD/kg for TAC and $1.329x10^8$ pts/kg for EI99.

The table delineates the complexities and interdependencies of various units in the process, highlighting the intricate balance required for efficient and sustainable operation.The Pretreatment and Simultaneous Saccharification and Fermentation (SSF) units are foundational in processing biomass. Pretreatment, with a volume of 625.98 m³ and a significant duty of 166.47 MW, indicates its role in preparing biomass for enzymatic hydrolysis, while SSF, with a larger volume of 8998.3 m³ and a duty of 2 MW, suggests its centrality in converting biomass to butanol. The enormous water and biomass flows through these units
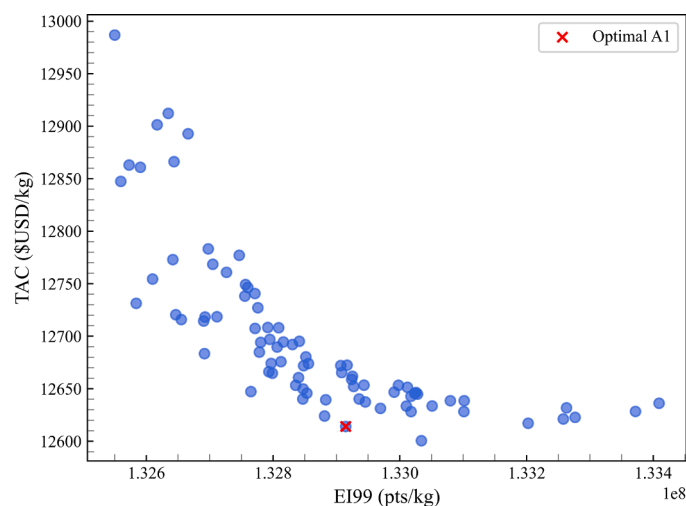
Table 4

Design parameters for biomass-butanol processes.

Parameter	Equipment					
	Pretreatment	*SSF*	*LLE*	*C1*	*Prefractionator*	*Petlyuk*
N_t	–	–	8	41	12	61
N_f	–	–	–	28	5	–
R_r	–	–	–	0.9098	–	5.5618
LIF_{IS}	–	–	–	–	–	15
VIF_{IS}	–	–	–	–	–	44
LIF_{OS}	–	–	–	–	–	15
VIF_{OS}	–	–	–	–	–	47
SPF_{OS}	–	–	–	–	–	29
LIF	–	–	–	–	–	23,802.8
VIF	–	–	–	–	–	50,449.3
SPF	–	–	–	–	–	13,787.8
D_r	–	–	–	145,487.9	–	40,169.8
V	625.98	8998.3	–	–	–	–
Q	166.47	2	–	2 698	–	43.22
W_F	462,632.5	3501,518.2	–	–	–	–
B_F	462,632.5	–	–	–	–	–
EA_F	–	–	2661,720.1	–	–	–
E_F	–	14,711.08	–	–	–	–

Fig. 10. Pareto front for biomass-biojet process.

(462,632.5 kg/h and 3501,518.2 kg/h, respectively) underscore their critical roles in the process. The Liquid-Liquid Extraction (LLE) unit, crucial for separation processes, is relatively smaller in scale but handles a massive extractant agent flow of 2661,720.1 kg/h. This high flow rate reflects the necessity for efficient separation in butanol production.

The C1 column, with 41 stages and a feed input at stage 28, demonstrates its complexity in the distillation process. A notable distillate rate of 145,487.9 kg/h and a reflux ratio of 0.9098 highlight its efficiency in product purification. The duty of 2698 MW for this unit indicates a significant energy input, emphasizing its operational significance. The Petlyuk column, with a remarkable 61 stages and multiple input and output stages, handles various streams, including LIFII, VIFII, and SPF, with flow rates of 23,802.8 kg/h, 50,449.3 kg/h, and 13,787.8 kg/h, respectively. Its distillate rate of 40,169.8 kg/h and duty of 43.22 MW further illustrate its complex role in refining and purifying the product.

The design obtained for the biomass-butanol process, implementing the LHW pretreatment and intensified reaction (SSF reactor) and separation (Petlyuk column) equipment, is shown in Table 4. It can be observed in the design that the column recovering the extractant agent (C1) consumes an excessive amount of energy in the reboiler duty. It is important to note that the design obtained for this process is capable of producing 91,358 kg/h of butanol, corresponding to the bottom stream of the Petlyuk column.

In the realm of sustainable aviation fuel production from butanol, a meticulous examination of the design parameters for butanol-biojet processes, as delineated in Table 5, provides a profound understanding of the process intricacies, reminiscent of the analytical approach espoused by chemical engineer Ignacio Grossmann. The C2 column, a critical component in the process, is characterized by a comprehensive set of parameters: it operates with 17 stages, with feed input at stage 15, an exceptionally high reflux ratio of 195.32, and an output stage of SP located at stage 8. The significance of these parameters becomes evident in the context of process efficiency and separation effectiveness, with the C2 column managing a substantial SPF (Stream Permeate Flow) of 45,615.18 kg/h and a distillate rate of 20,721.48 kg/h. This indicates a highly optimized separation process, crucial for achieving desired purity levels in the biojet fuel.

In contrast, the Dehydration, Oligomerization, and Hydrogenation units are characterized by distinct yet equally vital parameters. The Dehydration unit, with a volume of 3515.46 m^3 and a duty of 29.95 MW, indicates a large-scale operation with significant energy input, suggesting its critical role in removing water to achieve the desired butanol purity. The Oligomerization unit, slightly smaller in volume at 3369.95 m^3, does not have a specified duty, implying a different operational

focus, potentially on molecular rearrangement processes. The Hydrogenation unit, while modest in size (13.33 m^3), plays a pivotal role in the hydrogenation process, as indicated by a hydrogen flow of 555.02 kg/h and a duty of 14.59 MW, reflecting its importance in facilitating chemical reactions essential for producing the final biojet fuel. The biojet production is reflected in the side product of column C2. The obtained design can produce 45,833 kg/h of biojet from the butanol produced in the previous process.

In Figs. 11 and 12, the equipment contribution to the overall biomass-biojet process is shown for both objective functions, total annual cost, and eco-indicator 99. It is important to highlight that the equipment related to the biomass-butanol process contributes more significantly to both objective functions. In Table 6, a critical analysis of the equipment contributions to Total Annual Cost (TAC) and Environmental Impact (EI99) reveals pivotal insights for process intensification. The C1 unit emerges as a paramount factor, commanding a dominant 89.23 % of the TAC and a substantial 49.16 % in EI99, underscoring its pivotal role in economic viability and environmental footprint. In stark contrast, the Liquid-Liquid Extraction (LLE) unit, while minimally impacting the TAC at 0.02 %, significantly influences EI99 with a 43.49 % contribution, indicating a disproportionate environmental impact, possibly linked to solvent use or energy-intensive separation processes. The Simultaneous Saccharification and Fermentation (SSF) unit presents an intriguing disparity, being economically marginal (1.63 % of TAC) yet considerably impactful environmentally (5.92 % of EI99), suggesting a potential area for environmental optimization. The cumulative impact of ancillary units like Petlyuk, Dehydration, Oligomerization, Hydrogenation, and C2, though individually minor in both TAC and EI99, warrants attention for holistic process enhancement. comprehensive analysis, as delineated in Table 6, offers a critical evaluation of the economic and environmental implications of equipment used in the production of sustainable aviation fuel from butanol. Notably, the Pretreatment process, while accounting for a modest 3.38 % of the Total Annual Cost (TAC), has a relatively low environmental impact, contributing just 0.67 % to the EI99 metric. This contrasts sharply with the Liquid-Liquid Extraction (LLE) system, which, despite its negligible contribution to TAC (a mere 0.02 %), poses a substantial environmental burden, accounting for 43.49 % of EI99. The most economically significant equipment is C1, commanding an overwhelming 89.23 % of the TAC, while concurrently representing a significant environmental concern with a 49.16 % contribution to EI99. Other processes such as SSF, Petlyuk distillation, Dehydration, Oligomerization, and Hydrogenation show varied but generally lower impacts on both TAC and EI99. This detailed breakdown underscores the necessity for process intensification and optimization strategies that can effectively balance cost-efficiency with environmental sustainability in the realm of biofuel production.

5. Discussion

In this study, the differential evolution method augmented with a tabu list was employed to optimize the designs of both the intensified biomass-butanol process and the conventional butanol-biojet process. This optimization aimed to minimize the total annual cost (TAC) and environmental impact, as quantified by our selected metrics. Upon comparing the two processes, it became apparent that the biomass-butanol process exerts a more substantial influence on both the TAC and environmental metrics than the butanol-biojet process. The primary challenge within the biomass-butanol process is attributed to the equipment configuration dedicated to breaking the azeotropes in the separation liquid from the reaction stage. This issue is particularly evident in the columns for liquid-liquid extraction and the recovery of the extractant agent. This specific configuration accounts for a significant portion of the total cost and environmental impact, amounting to 89.25 % of the TAC and 92.65 % of the environmental impact, respectively.
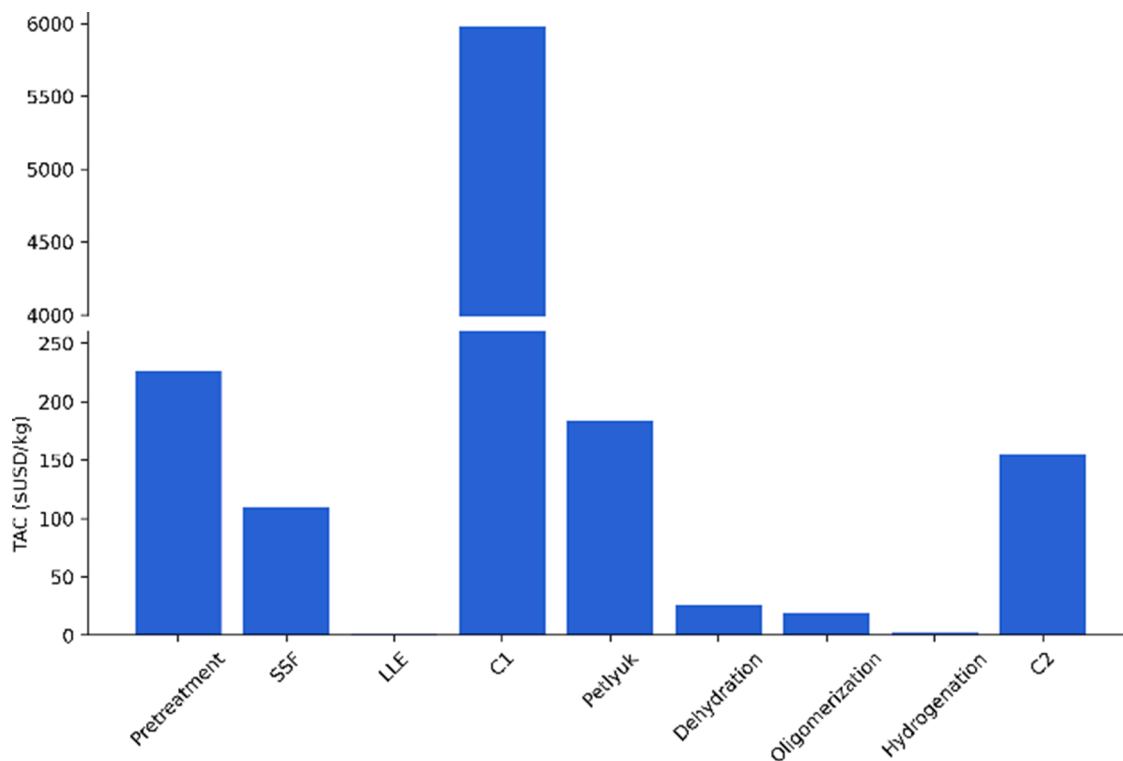
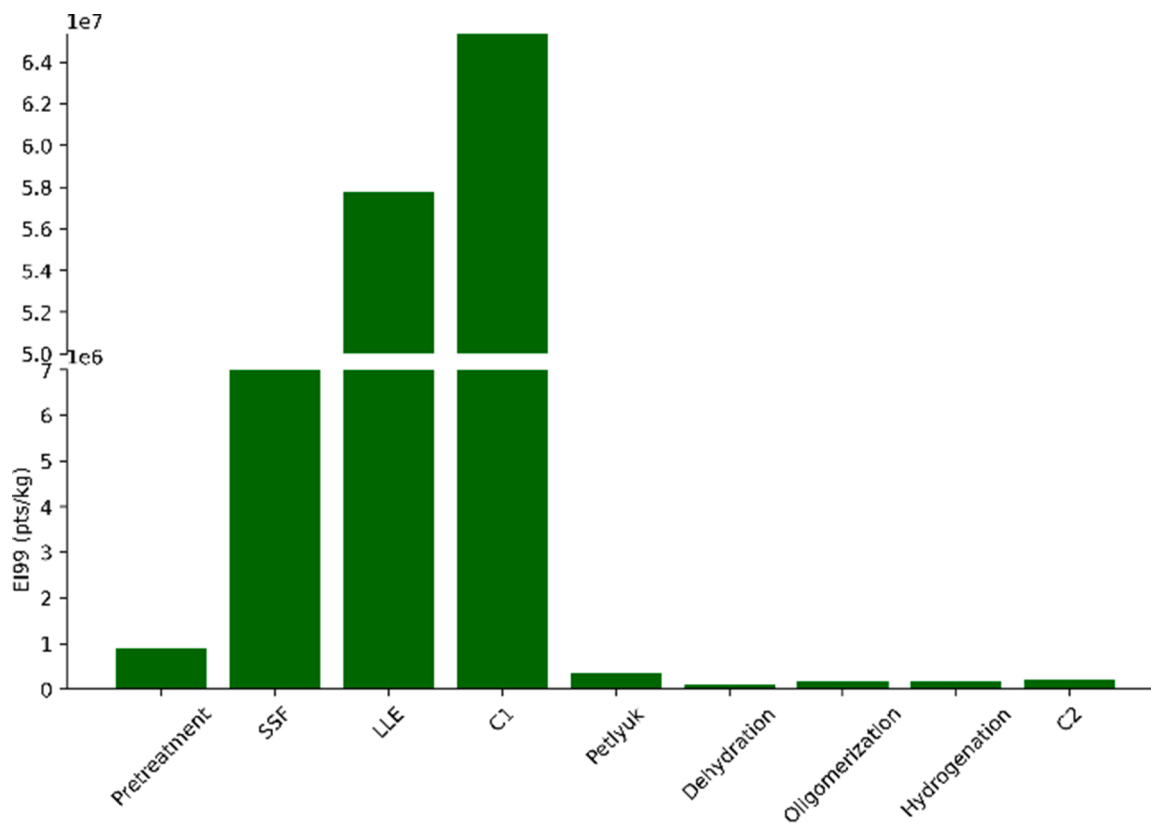**Fig. 11.** Contribution to the TAC per equipment.

Fig. 12. Contribution to the EI99 per equipment.

A critical evaluation against the United Nations' 2030 Agenda, specifically objectives 7 (Affordable and Clean Energy) and 12 (Responsible Consumption and Production), reveals that the biomass-biojet process design does not align with these goals. This misalignment is primarily due to its high environmental impact and elevated annual costs. Therefore, while this process contributes to the promotion of clean energy, it falls short in terms of affordability and sustainability. This observation underscores the necessity for further refinement in the

Table 6
Percentage contribution by equipment to objective functions.

Equipment	TAC	EI99
Pretreatment	3.38 %	0.67 %
SSF	1.63 %	5.92 %
LLE	0.02 %	43.49 %
C1	89.23 %	49.16 %
Petlyuk	2.75 %	0.27 %
Dehydration	0.39 %	0.07 %
Oligomerization	0.28 %	0.12 %
Hydrogenation	0.03 %	0.12 %
C2	2.30 %	0.17 %

process design, with a specific focus on developing more cost-effective and environmentally benign solutions for the separation stage. Future iterations of this process should aim to better align with sustainable development goals, ensuring that clean energy solutions are both economically viable and environmentally responsible.

In the biomass-butanol processes, parameters such as stages, feed input stage, reflux ratio, and various flow rates (including water, biomass, and extractant agent) were meticulously optimized. The focus on process intensification is evident in the optimization of stages and flow rates in the SSF, LLE, C1, and Petlyuk processes. The high volume of biomass and water flow, along with significant enzyme flow in the SSF process, underscores the scale and efficiency achieved. Similarly, the butanol-biojet processes show a streamlined approach in the dehydration, oligomerization, and hydrogenation stages, with particular attention to the C2 process. The design parameters like the number of stages, feed input stage, and distillate rate reveal a fine-tuned process aimed at maximizing output while maintaining efficiency. The significant volume and duty values in dehydration and oligomerization processes highlight the scale of operation, with the hydrogenation process optimizing hydrogen flow for effective fuel synthesis.

These results underscore the study's success in enhancing the economic and environmental viability of sustainable aviation fuel production. By focusing on process intensification and optimization across various stages, the study contributes to the development of more sustainable aviation fuel options, aligning with global efforts to reduce carbon emissions in the aviation sector. The detailed parameter optimization also suggests potential avenues for further research and development in the field, aiming to make sustainable aviation fuels a more accessible and efficient energy source for the future.

6. Conclusions

The modeling and simulation of the ATJ process for biojet fuel production, utilizing butanol, are detailed, a notable breakthrough has been achieved in the realm of renewable fuel generation, specifically through the strategic incorporation of advanced process intensification techniques within the butanol-biomass conversion pathway. These innovations, in tandem with the conventional Alcohol-to-Jet (ATJ) conversion methodology, have facilitated a markedly more efficient and robust framework for biofuel production. An intricate design for the biomass-to-biojet fuel conversion pathway was formulated through the application of a sophisticated stochastic optimization technique, blending differential evolution with a tabu list approach. This design underwent a thorough evaluation against stringent environmental and economic metrics, unveiling a TAC of USD 12,614/kg and an EI99 index of 1.329×10^8 points/kg. Impressively, this optimized process configuration demonstrated a substantial throughput, with a production yield of 91,358 kg/h of butanol and 45,833 kg/h of biojet fuel.

The findings of this study mark a significant milestone in the domain of sustainable fuel production, reflecting not only advancements but also highlighting the transformative potential of integrating intensification and process optimization strategies across economic and environmental dimensions. Through meticulous investigation, it has been elucidated

that the adoption of intensified methodologies holds considerable promise in bolstering the efficiency and sustainability of aviation biofuel production processes. By leveraging these intensified approaches, substantial improvements in operational cost reduction and environmental footprint mitigation are achievable, thereby underscoring the practical feasibility and tangible benefits of such strategies. Consequently, the implications of this research extend beyond mere academic discourse, offering compelling evidence and actionable insights that can inform industry stakeholders and policymakers alike in navigating towards a more sustainable and ecologically responsible trajectory for the aviation sector.

CRediT authorship contribution statement

Angel Eduardo García-Hernández: Writing – review & editing, Writing – original draft, Methodology, Investigation, Conceptualization. **Juan Gabriel Segovia-Hernández:** Writing – original draft, Supervision, Resources, Investigation, Conceptualization. **Eduardo Sánchez-Ramírez:** Writing – original draft, Supervision, Methodology, Investigation. **Gabriel Contreras Zarazúa:** Writing – review & editing, Writing – original draft, Methodology, Investigation. **Ivan Fernando Hernandez Araujo:** Writing – original draft, Validation, Methodology. **Juan José Quiroz-Ramírez:** Writing – review & editing, Visualization, Supervision, Investigation.

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.

Data availability

The authors are unable or have chosen not to specify which data has been used.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.cep.2024.109769.

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