

## Article

# Novel Heat-Integrated Hybrid Distillation and Adsorption Process for Coproduction of Cellulosic Ethanol, Heat, and Electricity from Actual Lignocellulosic Fermentation Broth

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**Abstract:** Cellulosic ethanol (CE) can not only be produced from a nonedible, cheap, and abundant lignocellulose feedstock but also can reduce carbon footprint significantly compared to starch ethanol. Despite great stimulation worldwide, CE production has not yet commercialized because of the complexity of lignocellulose. Therefore, intensive research and development are needed to improve CE technologies. In this study, a cost-efficient and sustainable design was proposed for the coproduction of CE, heat, and electricity from the actual lignocellulosic fermentation broth. First, a conventional coproduction process of CE, heat, and electricity based on hybrid distillation and adsorption (HDA) was simulated and optimized. Subsequently, various heat integrated (HI) techniques such as heat pump (HP), multi-effect distillation (MED), and combined HP-MED were evaluated to improve the CE process. The combined heat and power (CHP) process that utilized the combustible solids of the beer stillage was designed and integrated with the CE process. Structural alternatives were assessed for both economic and environmental impacts. The results show that the proposed HI-HDA process can save 36.9% and 33.6% of total annual costs and carbon footprint, respectively, compared to the conventional CE process. In the proposed HI-HDA coproduction process, the CE recovery process can be self-efficient in energy and the CHP can generate 12.0% more electricity than that in the conventional coproduction process.

**Keywords:** cellulosic ethanol; lignocellulosic biomass; hybrid distillation and absorption; process integration; coproduction



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## 1. Introduction

The current fossil-based economy poses several crucial problems, such as mineral-resource depletion and contribution to global warming. Thus, the transition to a renewable resource-based economy is essential for a sustainable future. Biomass, which is a carbon-neutral material, is the most promising candidate to replace fossil fuels. Particularly, lignocellulosic biomass is the most abundant renewable material on Earth and is a readily renewable resource for the industry in forms of softwood, energy crops, grasses, and agricultural residues [1].

Biofuels, such as bioethanol, biodiesels, and biogas that are produced from lignocellulosic biomass in increasing demands have the potential to drive a sustainable bioeconomy. Of these, bioethanol is one of the most important biofuels that can be mixed with gasoline for use in current engines without the need for engine modification to reduce carbon monoxide and other smog-causing emissions. The first generation of bioethanol (i.e., starch bioethanol) is mainly produced by the fermentation of sugars in food crops, such as corn and wheat, and is already commercialized, being used in the transportation sector worldwide. Although the technologies related to starch bioethanol are well developed, the price of starch feedstock is relatively high and accounts for 40–70% of the total production cost [2].

Another drawback of starch bioethanol is the associated food-versus-fuel tradeoff, arising from the increased food prices and the detrimental effect on food supplies associated with its production and use. Therefore, the production of cellulosic ethanol (CE) from lignocellulosic biomass, a nonedible, cheap, and abundant material, has been extensively explored. Lignocellulosic feedstock, such as corn stover ethanol, can reduce carbon dioxide (CO<sub>2</sub>) emissions by 37% and 86% more than corn ethanol and oil-based ethanol, respectively, resulting in the improved sustainability of bioethanol production [3,4].

However, the complex composition of lignocellulosic biomass, which consists of hemicellulose (20–35%), cellulose (30–55%), and lignin (10–25%), results in the requirement for a high degree of complexity in the processing and separation technologies. This causes the production cost of CE to be considerably higher than that of starch bioethanol, even though lignocellulose is substantially cheaper than starch crops [5]. Therefore, commercial-scale CE production is yet to begin, and further research and development activities are required. In recent years, several CE pilot and demonstration plants employing new cost-effective technologies have been launched [6]. In 2015, Beta Renewables started producing CE at its 40 million gallons per year (MMgy) plant in Crescentino, Italy [7]. However, it was sold to pay off debts in 2018 because of the bankruptcy of its parent company [8]. Abengoa's 25 MMgy CE plant located in Kansas, USA, began operation in 2015; however, they were also declared bankrupt after only one year of operation [7]. In 2016, DuPont celebrated the opening of a 30 MMgy CE plant in Nevada, USA. By 2017, the plant was under new ownership by Verbio Vereinigte BioEnergie AG [7]. In spite of these efforts, it is clear that creating a more competitive global CE market is challenging.

CE production typically consists of three steps: pretreatment, fermentation, and separation. First, the biomass feedstock undergoes a pretreatment step prior to fermentation and production of the beer fermentation broth. This broth, which contains 3–12 wt% ethanol, is then separated to give ethanol in commercially acceptable purity. A large amount of water present in the fermentation broth and the formation of a homogeneous azeotropic ethanol/water mixture are two of the main reasons that the separation step is energy intensive. Various separation techniques have been proposed to overcome the challenges of an azeotropic mixture, such as azeotropic distillation (AD), adsorption, membrane vapor permeation, membrane pervaporation, extractive distillation (ED), and pressure swing distillation [9–12]. Of these, the capacities of pervaporation and vapor permeation have reached their limits and are cost intensive on an industrial scale [13]. ED is more energy effective than AD and is widely used in industrial bioethanol production [9]. A dividing-wall column using extracting solvent ethylene glycol (EG) was reported to improve the energy requirement by 17% compared to the conventional process [14]. However, the use of EG as the extracting solvent may cause several serious environmental issues, so its use has been restricted in several countries [15]. Adsorption using molecular sieves has been reported to be an efficient way to overcome the ethanol/water azeotrope [10,16]. In particular, adsorption using molecular sieves is particularly popular for removing small amounts of water from an ethanol/water mixture because of its selectivity and ease of maintenance [12].

However, most studies on the ethanol separation process assume that the beer feed from the fermentation process is composed of only ethanol and water. This assumption results in a simpler process but does not represent the actual lignocellulosic composition. Breaking down the cellulose-hemicellulose-lignin structure can lead to the presence of more compounds in the fermentation output, resulting in the requirement for a more complex separation procedure [17,18]. Humbird et al. reported a cost-efficient process that converted lignocellulosic biomass to CE, considering a real composition of corn stover [10]. The biomass feedstock was converted to CE through dilute-acid pretreatment, enzymatic hydrolysis, and saccharification processes. The dilute CE stream was then fed to two distillation columns and one adsorption column using molecular sieve to reach commercial-grade ethanol (99.5 wt%). Based on the NREL (National Renewable Energy Laboratory) process, a coproduction process of ethanol and furfural was developed to

efficiently utilize both the cellulose and hemicellulose parts in lignocellulose materials [16]. In these processes, lignin and unconverted cellulose and hemicellulose were used for the production of heat and electricity through the biomass combined heat and power (CHP) process. In the biomass CHP process, the direct CO<sub>2</sub> emission can be close to zero because of the CO<sub>2</sub> emissions from biomass combustion having already been absorbed during plant growth [19]. Utilizing the solid byproducts to reduce the solid waste disposal costs is currently the critical issue in industrial biorefinery [20,21]. In the study, obtaining a high degree of integration and utilizing all parts of lignocellulose are key strategies for reducing CE production costs to make it more competitive with fossil fuels.

In this study, a novel heat-integrated (HI) hybrid distillation and adsorption (HI-HDA) process for the coproduction of CE, electricity, and heat from an actual lignocellulosic fermentation broth was proposed. First, a CE conventional process consisting of pre-concentration distillation and HDA was simulated and optimized based on NREL processes [10]. Subsequently, various HI techniques such as heat pump (HP), multiple-effect distillation (MED), and combined HP-MED were applied to improve the energy efficiency of the CE separation process. The combustion, steam boiler, and turbine generation (or CHP process) were then proposed to utilize a lignin fraction in the beer stillage from CE purification process. For a fair comparison, structural alternatives were assessed for both economic and environmental impacts. Consequently, promising design and operating conditions for the coproduction of CE, heat, and electricity were explored.

## 2. Methods

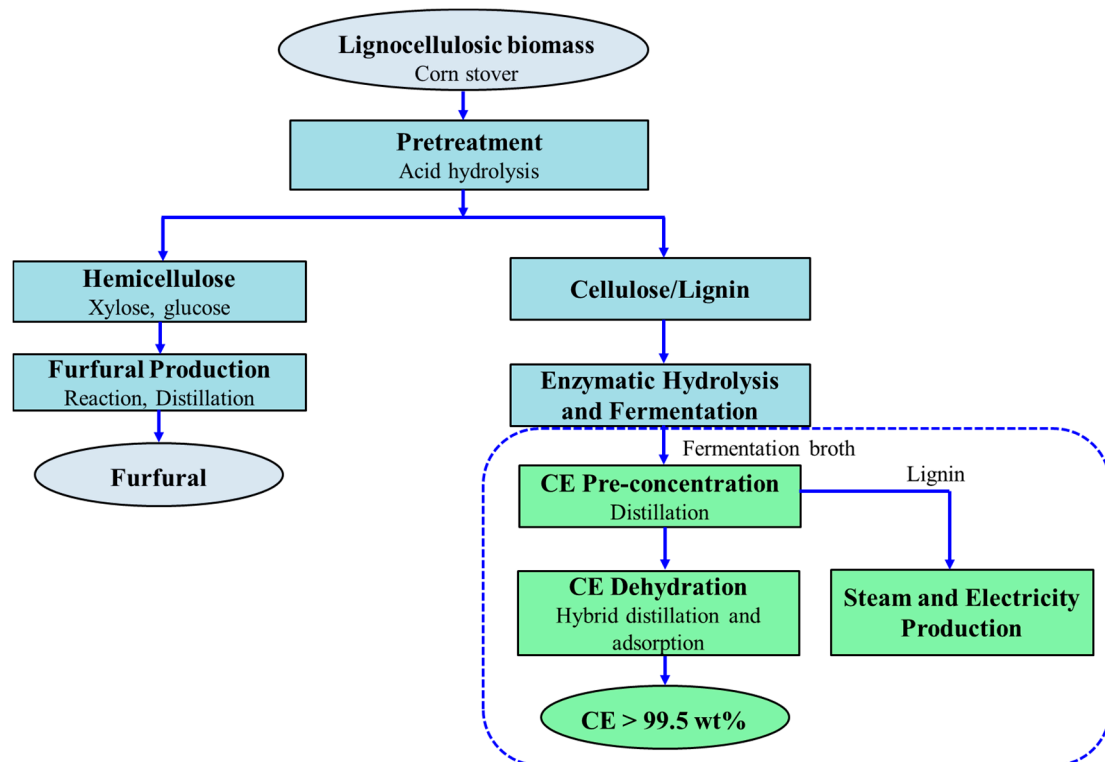
### 2.1. Design and Simulation

In this study, a process for the coproduction of CE, heat, and electricity was designed with a CE production capacity of 120 kilotons per year (ktpy). Table 1 lists the component compositions of the actual fermentation broth in detail. The broth comprises a number of components, including CE, water, gas (CO<sub>2</sub>), soluble solids, and insoluble solids. A conventional process was designed based on processes reported previously [10,16,22]. Figure 1 shows a schematic flowsheet of the integrated process for coproduction of CE, furfural, heat, and electricity from lignocellulosic feedstock. The process coproduces CE and furfural, and the concept was initially proposed by Strømsnes et al. [16]. In a previous study, a novel reactive distillation with an extraction-distillation configuration was proposed to improve the furfural production process [22]. In the present study, a cost-effective design for the coproduction of CE and electricity from a lignocellulosic fermentation broth (Figure 1, inside the dashed line rectangle) was developed. The lignocellulosic feedstock first undergoes a pretreatment step of acidic hydrolysis using sulfuric acid as the catalyst. In this step, hemicellulose was converted into its sugar monomers: mainly xylose and glucose. The xylose solution was separated from the cellulose/lignin slurry by a solid/liquid separation unit and delivered to the furfural production process. The details of the furfural production process design are beyond the scope of this study. The cellulose/lignin slurry was then introduced to the enzymatic-hydrolysis and fermentation processes in which CE was produced. The fermentation broth, which contained 3–12 wt% CE, was transferred into a distillation column to pre-concentrate CE to approximately 39 wt% as a vapor side-stream. This vapor stream was introduced to an HDA process to achieve a commercial purity of 99.5 wt%. The beer stillage from the bottom of the pre-concentration column was transferred to a solid/liquid separator to separate the lignin and the water. Wastewater was sent to a wastewater treatment process to produce methane via anaerobic digestion. Methane and combustible solids were then burned to produce steam, which was used to supply the heating demand in the plant and generate electricity.

**Table 1.** Feed mixture conditions and product specifications.

Component	Mass Fraction (wt%)
H <sub>2</sub> O	90.46%
Extractives *	0.21%
Dextrose	0.17%
Xylose *	0.04%
Ash *	0.90%
Lactic acid	0.05%
Furfural	0.01%
Vanillin	2.88%
Ethanol	3.16%
CO <sub>2</sub>	0.11%
Cellulose *	0.61%
Xylan *	0.36%
Ammonium sulphate *	0.06%
Diammonium phosphate *	0.03%
Protein *	0.75%
Cell mass *	0.14%
Ammonium acetate *	0.03%
Temperature (°C)	32
Pressure (atm)	1
Mass flowrate (kg/h)	455,247
CE purity (wt%)	99.5

\* insoluble solids.

**Figure 1.** Schematic flowsheet of the process for coproduction of furfural, CE, heat, and electricity from lignocellulosic biomass.

All processes were rigorously simulated using Aspen HYSYS V.10. The physical properties for all components were taken from the Aspen database and the NREL report [10]. The non-random two-liquid (NRTL) fluid package was used to calculate the liquid activity

coefficients. In addition, the UNIFAC method was used to estimate the missing binary parameters in the vapor-liquid equilibrium (VLE) data.

Several simulations were first run to optimize the distillation column and determine the initial structure. Subsequently, the feed location and total number of stages were adjusted to minimize the total annual cost (TAC) while maintaining the target product purity and recovery. The minimized TAC provided an optimal tradeoff between capital and operating costs; accordingly the optimal operating conditions and design were explored. The detailed optimization procedure for a distillation column was reported previously [23].

## 2.2. Economic Evaluation

For a fair comparison of the economic impact, the total investment cost (TIC), total operating cost (TOC), and TAC of all process alternatives were estimated, as described in previous studies [23]. Equipment costs were estimated using the correlations from Turton et al. and Biegler et al. [24,25]. The tray sizing function in Aspen HYSYS was used to estimate tray spacing, column heights, and the column diameters. A Chemical Engineering Index of 596.2 (corresponding to 2020) was used to update the TIC estimates. The heat exchangers, compressors, condensers, reboilers, tray stacks, column vessels, and blowers were considered in the TIC. A cooling water price of 0.35 \$/GJ, low-pressure steam price of 13.28 \$/GJ, and high-pressure steam of 17.70 \$/GJ were used for the TOC calculations [25]. A plant lifetime of 10 years was assumed while a fixed interest rate of 8% was used for the TAC estimates.

## 2.3. Environmental Assessment

The total annual CO<sub>2</sub> emissions (TCE) of all processes were calculated for environmental assessment. Gadalla's method was used to calculate the CO<sub>2</sub> emissions for steam reboilers and compressors [26]:

$$[\text{CO}_2]_{\text{emiss}} = \left( \frac{Q_{\text{fuel}}}{\text{NHV}} \right) \left( \frac{C\%}{100} \right) \alpha, \quad (1)$$

where  $\text{NHV}$  is the net heating value of the fuel, and  $C\%$  is the carbon content. The molar mass ratio of CO<sub>2</sub> to C was  $\alpha = 3.67$ . Besides,  $Q_{\text{fuel}}$ , which denotes the amount of fuel used is calculated as below:

$$Q_{\text{fuel}} = \left( \frac{Q_{\text{proc}}}{\lambda_{\text{proc}}} \right) (h_{\text{proc}} - 419) \left( \frac{T_{\text{FTB}} - T_0}{T_{\text{FTB}} - T_{\text{stack}}} \right), \quad (2)$$

where  $Q_{\text{proc}}$  is the required heat duty of the system,  $\lambda_{\text{proc}}$  (kJ/kg) is the latent heat, and  $h_{\text{proc}}$  (kJ/kg) is enthalpy of steam. The flame temperature ( $T_{\text{FTB}}$ ) is 1800 °C, stack temperature ( $T_{\text{stack}}$ ) is 160 °C, and ambient temperature ( $T_0$ ) is and 25 °C.

## 3. Results and Discussion

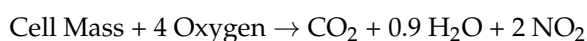
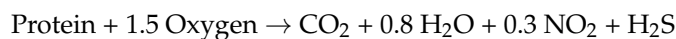
In this study, the CE plant was designed with a capacity of 120 ktpy of CE based on processes reported previously [10,16,22]. A conventional process for the coproduction of CE, heat, and electricity was designed and operating conditions were optimized. Several HI techniques, such as mechanical vapor recompression (MVR), MED, and combined MVR-MED, were applied to improve the CE recovery process. All the processes were evaluated for their economic and environmental impacts. The most promising design for the CE recovery process was found. The CHP process utilizing combustible solids in the beer stillage from the CE process was designed to supply heating and power demands of the plant. Finally, a cost-effective design with detailed design parameters and operating conditions was proposed for the coproduction of CE, heat, and electricity from an actual lignocellulosic fermentation broth.





The C1 bottom stream comprised a residual solid material (lignin, xylan, and cellulose) in water. A forced circulation reboiler was proposed to accommodate solids at the bottom of the column [10]. The insoluble solids were less than 4 wt% and were assumed to be highly porous and so nominal liquid flow was assumed. The beer stillage was heat-exchanged with the beer feed to 43 °C, which is a suitable temperature for liquid/solid separation. This cooled stream was introduced into a pressure filter to produce a solid cake, which was subsequently dried with air to 40% moisture. The filter was simulated as a component splitter with a split factor of 1.0 and 0.05, for solid and liquid components, respectively [10]. The wastewater was sent to the wastewater treatment process for anaerobic digestion, while the lignin cake was delivered to a burner. Although the wastewater treatment process was not simulated in this study, the amounts of methane and CO<sub>2</sub> produced by anaerobic digestion of organic components in the wastewater were calculated and used as raw materials for combustion. The calculation is based on the combined chemical oxygen demand (COD) for the organic matter present in wastewater. The detailed calculation was included in Appendix A.

The biogas (methane/CO<sub>2</sub>) was mixed with the air and preheated by the flue gas before being introduced into the burner. The burner was simulated as a conversion reactor in Aspen Plus, which assumes that all combustible material burns in oxygen with 100% conversion. The detailed reactions are as follows:



On the other hand, water fed to the heat exchanger circuit in the combustor was boiled and superheated to a high-pressure (HP) steam (435 °C, 50 atm). This HP steam was used to drive the primary turbine (T1), which turns a generator produced electricity for the plant. The isentropic efficiency of T1 were designed at 85%. The low-pressure (LP) steam (235 °C, 9.5 atm) from turbine T1 was then split into three steams for different uses in the process. Furthermore, 35 wt% of the LP steam was used to supply heating demand of C1, C2 reboilers and heater HT1 in the CE recovery process. For maximizing energy conversion, 56 wt% of the LP steam was used to drive the turbine T2 as steam was taken down to a vacuum pressure of 0.1 atm. The steam turbine T2 also produced electricity with the isentropic efficiency of 85%. The T2 output stream was condensed with cooling water and pressurized to 9.5 atm. In addition, 11 wt% of the LP steam was used to preheat the boiler feed water before being mixed with the condensate from the T2 output. The split ratio was adjusted to equal the heating demand of the plant, whereas excess electricity can be sold back to the grid. Furthermore, 20.79 MW and 20.14 MW of electricity were generated from the T1, T2, respectively, which corresponds to a cycle energy efficiency of 21.9%. All stream information including temperature, pressure, vapor fraction, and component composition is listed in Table 2.





Table 2. Cont.

Stream	1	2	3	4	5	6	7	8	9	10	11	12	13
CO <sub>2</sub>	0.002	0.002	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Cellulose	0.000	0.000	0.007	0.007	0.061	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Xylan *	0.000	0.000	0.004	0.004	0.036	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Ammonium sulphate *	0.000	0.000	0.001	0.001	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Protein *	0.000	0.000	0.008	0.008	0.075	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Cell Mass *	0.000	0.000	0.002	0.002	0.014	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Oxygen	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Nitrogen	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Methane	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
<b>Stream</b>	<b>27</b>	<b>28</b>	<b>29</b>	<b>30</b>	<b>31</b>	<b>32</b>	<b>33</b>	<b>34</b>	<b>35</b>	<b>36</b>			
<b>Vapor fraction</b>	0.0	0.0	0.0	0.0	0.0	0.0	1.0	1.0	1.0	0.0			
<b>Temperature (°C)</b>	82	138	125	128	132	289	175	35	25	89			
<b>Pressure (atm)</b>	9.5	9.5	9.5	9.5	50.0	1.0	1.0	1.0	1.0	1.5			
<b>Mass flowrate (kg/h)</b>	206,034	18,556	71,920	90,476	206,034	340,687	340,687	15,649	280,170	14,317			
<b>Mass fraction</b>													
Water	1.000	1.000	1.000	1.000	1.000	1.000	1.000	0.000	0.000	0.005			
Extractives *	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Dextrose	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Ash*	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Lactic acid	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Vanillin	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Ethanol	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.995			
Ammonia	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
CO <sub>2</sub>	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.741	0.000	0.000			
Cellulose	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Xylan *	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Ammonium sulphate *	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Protein *	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Cell mass *	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000			
Oxygen	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.233	0.000			
Nitrogen	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.767	0.000			
Methane	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.259	0.000	0.000			

\* insoluble solids.

Figure 1 shows the key design parameters of the optimized conventional coproduction process. Table 3 lists the key design parameters and hydraulics of all distillation columns used in the conventional process. The results show that C1 requires energy of 43,600 kW and 18,570 kW in the reboiler and condenser, respectively, whereas C2 requires 2269 kW and 14,570 kW in the reboiler and condenser, respectively. The high CE recovery of 99 wt% results in the relatively high energy usage in the C1 reboiler, which accounts for the tradeoff between CE loss and energy requirements. LP steam was used for all reboilers in the conventional process.

**Table 3.** Design parameters of all distillation columns.

	C1	C2
Tray type	Valve (Ballast-V1)	Valve (Ballast-V1)
Column diameter (m)	3.4	2.8
Number of trays	32	45
Number of flow paths	1	1
Max flooding (%)	85.5	85.0
Tray spacing (m)	0.61	0.61

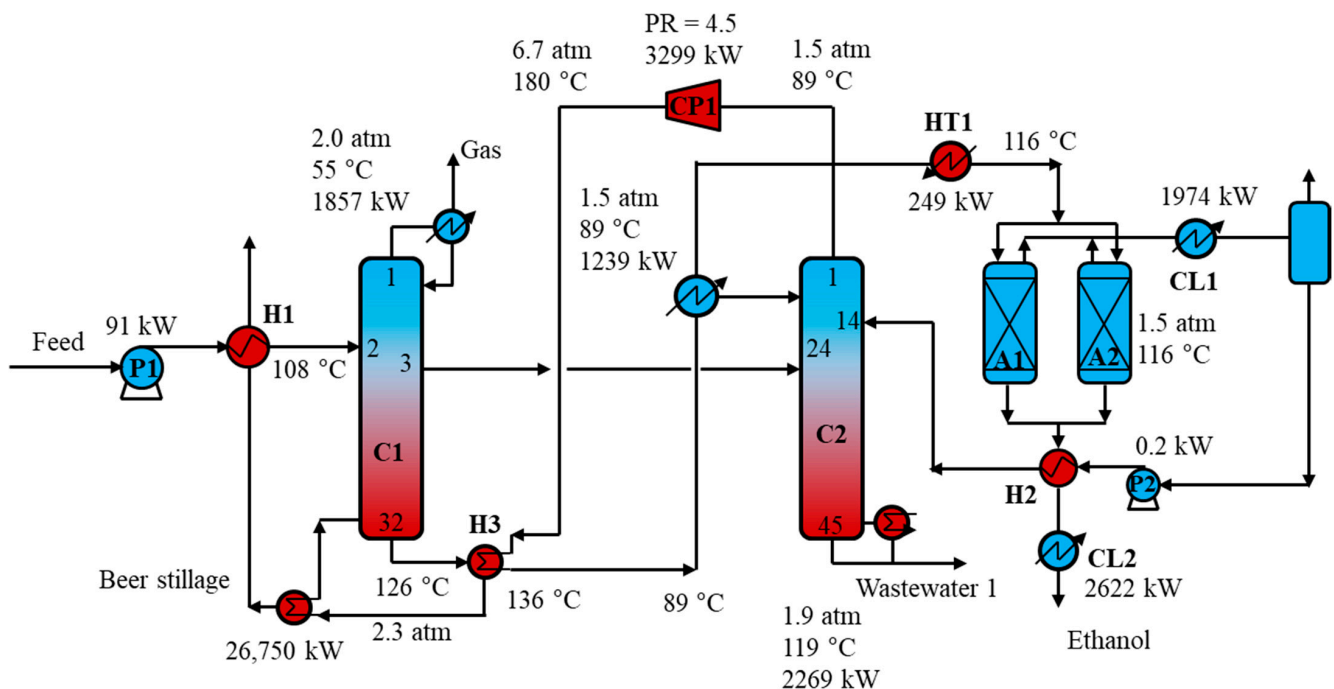
### 3.2. Improvement of the Coproduction Process of CE, Heat, and Electricity

Applying an appropriate heat-integrated technique is crucial for maximizing heat recovery in any chemical/biochemical process. Although distillation is the most common separation technology in chemical/biochemical processes, it has a relatively low thermodynamic efficiency. A distillation column often consumes a large amount of energy at high temperatures in the reboiler, whereas a similar amount of energy is released at low temperatures in the condenser [27]. As shown in Figure 1, heat is rejected at the C1 condenser, C2 condenser, and cooler CL1, whereas a large amount of energy is required in the C1 reboiler, C2 reboiler, and heater HT1. Therefore, there was an opportunity to make an energy integration to utilize the heat sources (condensers) to supply the heat sink (reboilers), resulting in a decrease in the energy requirement of the entire process.

#### 3.2.1. HP-Integrated HDA of the CE Process

HP are state-of-the-art systems that can improve the energy efficiency of conventional distillation by transfer heat from a lower temperature at a condenser to a higher temperature at a reboiler. Several HP concepts such as MVR, vapor compression, and thermal vapor recompression have been proposed to upgrade the discharge energy in the condenser to reduce the consumed valuable utilities [27]. Of these, MVR, which is an energy-efficient system for binary distillation, has been widely applied in industrial separation processes [27]. Figure 3 shows a schematic diagram of the MVR configuration of the CE process with the main design parameters. First, the top vapor from C2, which was used as the heat transfer medium to supply heat to the C1 reboiler, was pressurized from 1.5 atm to 6.7 atm at 180 °C by compressor CP1. The pressure ratio of CP1 was designed to obtain a minimum temperature difference of 10 °C in heat exchanger H3. The stream from C1 going to the reboiler was heated by the compressor output through heat exchanger H3 and fed to the C1 reboiler to obtain the target temperature. The C1 reboiler produced two outlet streams: the vapor stream was recycled back to C1, and the liquid stream was used to preheat the beer feed before going to the lignin combustion part. After transferring heat to the H3 cold inlet, the hot outlet of H3 (170 °C) was depressurized and fed to the C2 condenser to be partially condensed at 89 °C. The liquid output of the C2 condenser was refluxed to C2, whereas the vapor from the C2 condenser was heated further to 116 °C before being introduced into the adsorption column. The results show that the HP-assisted HDA process can save up to 72.4% and 36.5% of energy requirements in the condensers and reboilers, respectively, compared to the conventional CE production process. However, a multistage compressor (pressure ratio of 4.5) was required to pressurize the top vapor from 1.5 atm at 89 °C to 6.7 atm at 180 °C for safe compressor operation. The disadvantages

of using an HP process are the high investment costs and the process complexity associated with using a multistage compressor. Considering the sustainability of the process, the lower efficiency of the electricity source compared to the heat source results in higher CO<sub>2</sub> emissions in a compressor than in a reboiler with the same power [28]. The results show that the HP process can reduce the energy requirements of reboilers and condensers by 36.5% and 72.4%, respectively, compared to the conventional CE production process. However, the TIC of the HP process is 70.7% higher than that of the CE base case. Overall, the HP-integrated HAD process can save 28.9% and 21.0% of the TAC and TCE, respectively, compared to the conventional CE production process.

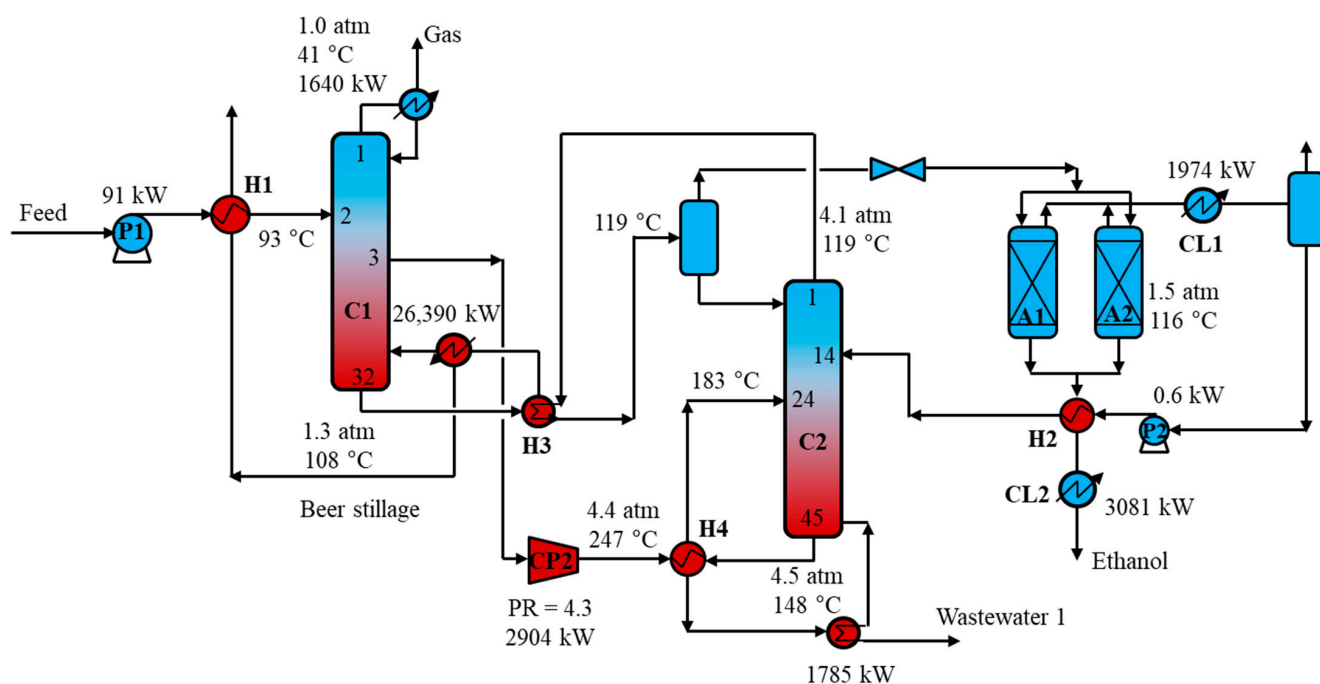


**Figure 3.** Schematic diagram of HP-integrated HDA for the CE production process from lignocellulosic fermentation broth. P1–2: pump; H1–2: heat exchangers; CP1: compressor; CL1–2: coolers; HT1: heaters; C1–2: distillation columns; A1–2: adsorption columns.

### 3.2.2. MED-Integrated HDA of the CE Process

MED consisting of two distillation columns, one operating at high pressure and the other operating at low pressure, is derived from a particular variant of heat integration. In this arrangement, a condenser of a high-pressure column acts as a reboiler of a low-pressure column [27]. Consequently, MED can utilize the rejected heat in the condenser of the high-pressure column to supply heat to the reboiler of the low-pressure column without using a compressor. Figure 4 depicts the schematic diagram of the MED-integrated HDA for the CE production process investigated in this study with key design parameters included. Compared to the conventional process, the first distillation column had a lower pressure of 1.3 atm, whereas the second column had a higher pressure of 4.5 atm. The operating pressures of the two columns were designed to obtain a temperature difference of 10 °C between the C2 top vapor and C1 bottom stream. In particular, the C1 bottom stream received heat from the C1 top vapor through heat exchanger H3 before being introduced to the C1 reboiler. Upon boiling of the stream in the reboiler, two new streams were produced. Of these, the vapor stream was recycled back to C1, and the liquid stream was used to preheat the beer feed before being sent to the CHP process. On the other hand, after transferring heat to the C1 bottom stream, the C2 top vapor stream was split into two streams: a liquid stream which was recycled to C2 in the first stage and a remaining vapor stream which was depressurized and input into the adsorption system. However,

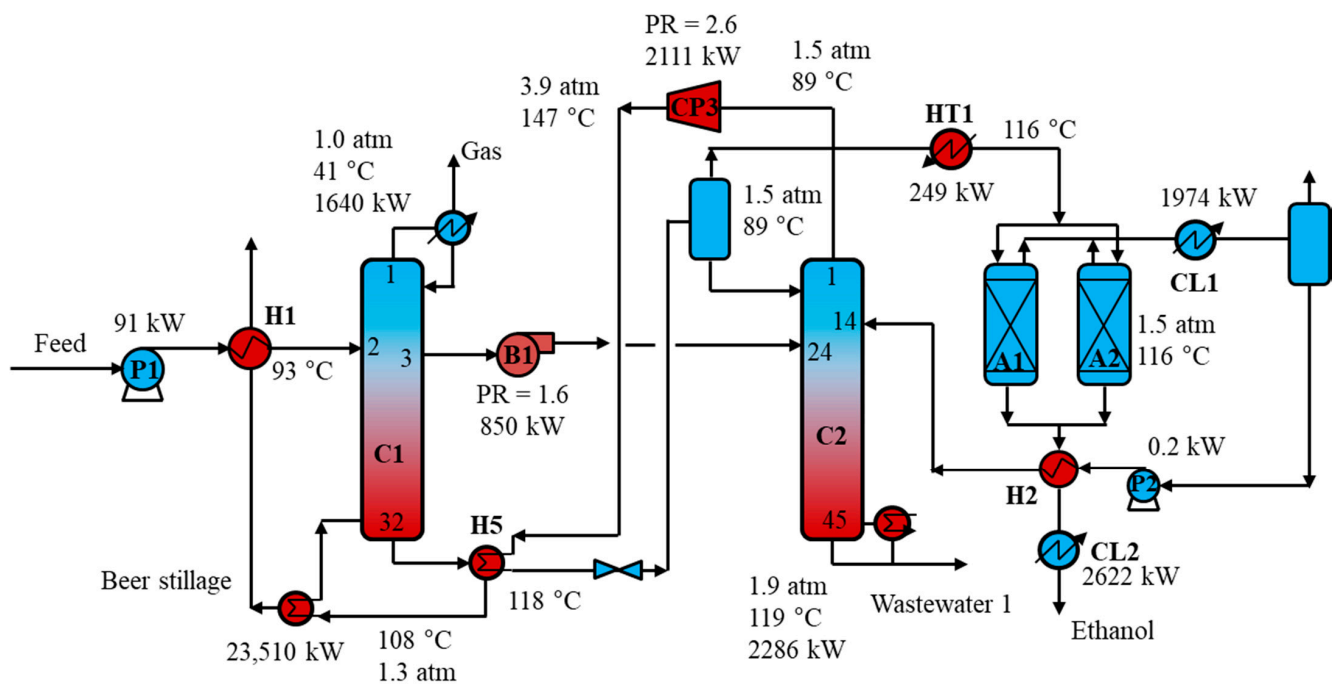
because C1 had a lower pressure than C2, a compressor was required to pressurize the side vapor stream of C1 before it was fed into C2. The C1 side vapor stream was pressurized from 1.0 atm to 4.4 atm before being introduced to the C2. The compressor CP2 has a relatively high power of 2904 kW and a pressure ratio of 4.4, which requires a multistage compressor instead of a one-stage compressor. The cost of investment in such a system is high and it introduces significant complexity in operation. Furthermore, the discharge temperature of 247 °C can cause compressor overheating and corrosion problems, which are currently serious issues of using the compressor in industry [29]. The results show that the MED-integrated HDA process can save up to 80.4% and 38.9% of energy requirements in the condensers and reboilers, respectively, compared to the conventional CE production process. Although the TIC of the MED process was 66.9% higher than that of the CE base case, the MED-integrated HDA process can save 31.4% and 25.2% of the TAC and TCE, respectively, compared to the conventional CE production process.



**Figure 4.** Schematic diagram of MED-integrated HAD for the CE production process from lignocellulosic fermentation broth. P1–2: pump; H1–4: heat exchangers; CP2: compressor; CL1–2: coolers; C1–2: distillation columns; A1–2: adsorption columns.

### 3.2.3. HI-HAD of the CE Process

Although the HP and MED processes described above show great performance in terms of energy efficiency, the use of multistage compressors is rather unstable and requires complex operation. Therefore, a heat-integrated HDA (HI-HDA) process that combines HP and MED was investigated. Figure 5 shows a schematic diagram of the HI-HDA process for CE production with key design parameters included. Compared to the conventional process, the C1 distillation column had a lower pressure, whereas the C2 pressure was kept constant. A heat pump with a compressor was integrated to utilize the heat source in the C2 condenser to supply heat to the C1 reboiler. Lowering the C1 pressure resulted in the need for a low-pressure ratio of compressor CP3. In particular, the C2 top vapor was pressurized from 1.5 atm at 89 °C to 3.9 atm at 147 °C, such that a one-stage compressor with a pressure ratio of 2.6 was sufficient. The C1 bottom stream was heated by the CP3 outlet and fed into the C1 reboiler. The C1 reboiler produced two streams: a vapor stream which was recycled back to C1 and a liquid stream which was used to preheat the beer feed before being introduced to the combustion part.



**Figure 5.** Schematic diagram of HI-HDA for the CE production process from lignocellulosic fermentation broth. P1–2: pump; H1–5: heat exchangers; CP3: compressor; CL1–2: coolers; HT1: heater; C1–2: distillation columns; A1–2: adsorption columns.

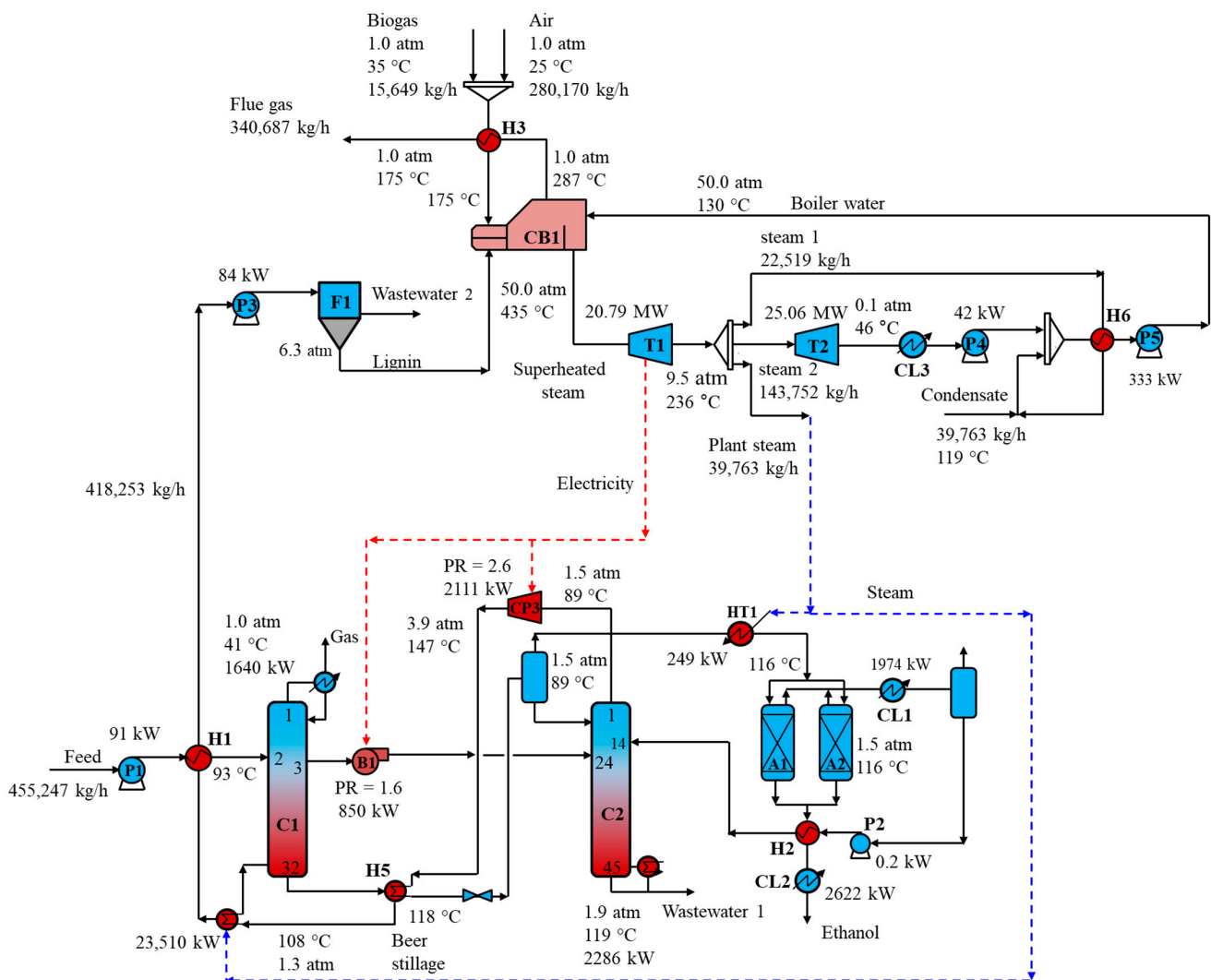
In addition, the side vapor of C1 was pressurized from 1.0 atm to 1.6 atm before being fed to C2. In this case, a blower with a pressure ratio of 1.6 was sufficient. The hot outlet of heat exchanger H5 was depressurized to 1.5 atm before being split into two streams: a liquid stream which was refluxed to C2 and a vapor stream which was heated to 116 °C before being fed into the adsorption column. Table 4 lists the key results of all structural alternatives for CE production process including energy requirements, production costs, and total amount of CO<sub>2</sub> emissions. The results show that the HI-HDA process can save up to 80.4% and 43.5% of energy requirements in the condensers and reboilers, respectively, compared to the conventional CE production process. Although the TIC of the HI process was 50.6% higher than that of the CE base case, the HI-HDA process can save 36.9% and 33.6% of TAC and TCE, respectively, compared to the conventional CE production process.

**Table 4.** Comparison of different structural alternatives for CE production process.

Structural Alternative	Conventional Process	HP Process	MED Process	HI Process
Reboiler duties (kW)	46,118	29,268	28,175	26,045
Reboiler duty savings (%)		36.5%	38.9%	43.5%
Condenser duties (kW)	18,401	5072	3614	3614
Condenser duty savings (%)		72.4%	80.4%	80.4%
Total investment costs (US k\$)	10,121	17271	16889	15246
Total investment cost savings (%)		−70.7%	−66.9%	−50.6%
Total operating costs (US k\$/year)	18,743	11,823	11,367	10,511
Total operating cost savings (%)		36.9%	39.4%	43.9%
Total annual costs (US k\$/year)	20,252	14,397	13,884	12,783
Total annual cost savings (%)		28.9%	31.4%	36.9%
Total carbon emissions (ton/year)	94,071	74,306	70,348	62,472
Total carbon emission reduction (%)		21.0%	25.2%	33.6%

### 3.2.4. Proposed HI-HDA Process for Coproduction of CE, Heat, Electricity

Figure 6 shows the key design parameters of the proposed HI-HDA process for coproduction of CE, heat, and electricity from fermentation broth. The HI-HDA configuration was proposed for improving the CE process while the turbogenerator was redesigned to match the steam demand of the CE process. In particular, the superheated steam was used to drive the turbine T1 to become the LP steam (9.5 atm, 236 °C) before being split into three streams. 19 wt% of the LP steam was used for the C1, C2 reboilers and heater HT1. 70 wt% of the LP steam was used to drive the turbine T2 to produce electricity with the isentropic efficiency of 85%. The T2 outlet at 0.1 atm and 46 °C was then condensed and pumped back to the boiler. The rest of the steam (11 wt%) was used to preheat the boiler feed water before it was fed to the boiler. The results showed that the CHP process could supply sufficient heat and power demand for the CE production process. In addition, 45.85 MW electricity was generated for selling back to the grid. The proposed HI-HDA coproduction process can increase 12.0% of electricity with respect to the conventional coproduction process. Note that the CE recovery process was thermal neutral in both conventional and proposed coproduction processes.



**Figure 6.** Schematic diagram of proposed HI-HDA process for coproduction of CE, heat, and electricity from lignocellulosic fermentation broth. P1–5: Pumps; H1–5: heat exchangers; C1–2: distillation columns; A1–2: adsorption columns; HT1: heater; CL1–3: coolers; T1–2: turbines; CB1: combustor + boiler; F1: pressure filter; CP3: compressor; B1: blower.



From the actual fermentation broth, a cost-efficient design for the coproduction of CE, heat, and electricity were developed in this study. It was found that utilizing the lignin fraction of lignocellulosic biomass means attaining a higher certain degree of integration in a biorefinery context.

#### 4. Conclusions

In this study, a cost-efficient and sustainable design was proposed for coproduction of CE, heat, and electricity from the actual lignocellulosic fermentation broth. The conventional coproduction process was successfully designed and optimized to achieve the targets. Various HI techniques such as HP, MED, and combined HP-MED demonstrated as attractive solutions to improve the conventional CE process. In particular, the results show that the proposed HI-HDA process can save 36.9% and 33.6% of TAC and CO<sub>2</sub> emission, respectively, compared to the CE conventional process. The biomass CHP process were efficiently integrated with the CE production process. The CHP process, which utilized the lignin and unconverted cellulose and hemicellulose, supplied sufficient steam and power for the CE recovery process to make the CE plant become thermal neutral. The proposed HI-HDA coproduction process can generate 12.0% more electricity than the conventional coproduction process. Furthermore, the solid waste disposal costs were reduced substantially and additional profit was generated from selling excess electricity back to the grid. The proposed coproduction process can be proposed both for constructing a new CE plant and a retrofit project which requires a short modification time. The results of this study provide a strong basis for the design and improvement of more sustainable CE production technologies from lignocellulosic biomass.

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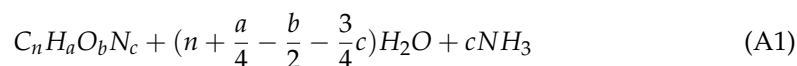
#### Abbreviations

AD	azeotropic distillation
CE	cellulosic ethanol
CHP	combined heat and power
COD	chemical oxygen demand
CO <sub>2</sub>	carbon dioxide
ED	extractive distillation
EG	ethylene glycol
HDA	hybrid distillation adsorption
HI	heat integrated
HP	heat pump
LP	low pressure
MED	multiple-effect distillation
MMgy	million gallons per year
MVR	mechanical vapor recompression
NHV	net heating value
NREL	National Renewable Energy Laboratory

TAC	total annual cost
TIC	total investment cost
TOC	total operating cost
UNIFAC	UNIQUAC functional-group activity coefficients
VLE	vapor-liquid equilibrium

## Appendix A Chemical Oxygen Demand (COD)

Equation of the oxidation of an arbitrary organic compound [16]:



Equation of mass COD [16]:

$$COD_m = \left(n + \frac{a}{4} - \frac{b}{2} - \frac{3}{4}c\right) \frac{M_w(O_2)}{M_w(C_nH_aO_bN_c)} \quad (A2)$$

**Table A1.** Calculated mass COD for compounds in the wastewater.

Compound	COD <sub>m</sub>
Lactic acid	1.065
Levulinic acid	1.516
Furfural	1.665
Acetic acid	1.067
Formic acid	0.348
Extractives	1.065
Glucose	1.065
Xylose	1.067

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