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## **HIGHLIGHTS**

- A sawdust mixture feedstock-based biorefinery producing bioethanol was developed
- The model was based our experimental database published in bioresource technology
- The plant converts 70,088 tons/year of biomass into 11,400 tons/year of ethanol
- Pinch Analysis lead to 19.1 and 18.7 MW of hot and cold utility, respectively
- Lignin, furfural, and other chemical production might reduce overall process costs

# 1 **Modeling and simulation of a sawdust mixture-based integrated biorefinery plant** 2 **producing bioethanol**

3

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6 Ferrand, France.

## 7 **Abstract**

8 The design, modeling and simulation of an integrated biorefinery plant assumed to convert  
9 different forestry assortments such as sawdust or shavings (sawmill waste) into bioethanol as  
10 the main product, lignin as the most valuable by-product, pellets as solid residue, and other  
11 valuable byproducts, was carried out. The proposed cellulosic ethanol biorefinery plant was  
12 simulated with ProSimPlus. The model was based on an experimental database involving the  
13 physicochemical characterization of the raw material, an Organosolv pretreatment as the  
14 deconstruction method, and enzymatic analysis. The investigated plant size processed 70,088  
15 tons of biomass/year, with a production capacity of 12,000 tons ethanol/year. Ethanol  
16 productivity reached 349 L/ton of dry feedstock. Considering water consumption,  
17 approximately 4.8 liters of water were needed to produce a liter of ethanol. Finally, the energy  
18 targeting through conventional Pinch Analysis lead to 19.1 MW and 18.7 MW of hot and cold  
19 utility energy demand for the entire process, respectively.

20 **Keywords:** Bioethanol; biorefinery modeling and simulation; biomass pretreatment;  
21 enzymatic hydrolysis; integrated biorefinery

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## 1. Introduction

Nowadays, the transition from the petroleum economy towards biofuel economy is highly accepted on both the national and international levels (Mesfun et al., 2019). Consequently, biofuels, especially from the second-generation, have gained a huge interest in the view of the current environmental problems and oil dependence. Furthermore, lignocellulosic biomass is perceived as an attractive candidate for second-generation biofuel production because its price is estimated as being the lowest compared to starch-based feedstock currently used to produce certain biofuels, and also because it is non-competitive with agro-food cultures (Bryngemark, 2019; Chovau et al. 2013).

France is endowed with forestry that can potentially provide a sustainable source of lignocellulosic biomass for bioethanol production. According to the national forest inventory, the forested mainland in France covers  $17.10^6$  ha. This corresponds to an afforestation rate of 29.7% of the territory close to the world rate and which is also increasing. It includes 136 species of trees (Rametsteiner et al., 2007). Therefore, the local wood processing industries generate a significant amount of multisource sawdust and other wastes, especially from softwood. However, if these multisource wastes were to be converted into biofuels, they could allow satisfying a wide part of energy demand, which is still mainly covered by petroleum products. Moreover, this will create a new opportunity for the forestry sector to contribute to the development of a reliable and sustainable national energy model that fits well with the energy transition policy and the fight against climate change (Sarks et al., 2014).

Sawdust from the sawmill industry is a low-cost industrial residue that contains a significant amount of carbohydrates with the potential to be a raw substrate for bioethanol production in a biorefinery plant. Its use could promote local valorization of wood waste with a view of setting up a forest biorefinery. Its chemical structure is mainly constituted of cellulose, hemicellulose, and lignin, which makes it recalcitrant to deconstruct, thereby limiting enzyme

1 ability to convert cellulose and hemicelluloses into sugars (Mupondwa et al., 2017). Hence, in  
2 order to make sugars available for bioconversion, four key steps are needed. First, the  
3 *pretreatment process* is compulsory to overcome the barrier that lignin constituted in the  
4 substrate. Thus, this step allows the destruction of the sawdust matrix to make the cellulose  
5 more accessible to the enzymes. Afterward, the pretreated solid is either *chemically* or  
6 *enzymatically hydrolyzed* to transform cellulose and hemicelluloses into sugars. Third,  
7 *fermentation* is performed on the rich sugar hydrolysate from the previous step to transform  
8 the latter into ethanol, and finally, *distillation* allows the separation of ethanol from the  
9 fermentation broth.

10 The pretreatment stage has been identified as the real technological and economic barrier to  
11 the development of lignocellulosic-based bioprocesses. Efficient pretreatment step is,  
12 therefore, essential to fractionate lignocellulosic structures for subsequent chemical  
13 transformations. An efficient pretreatment process must be able to reduce the crystallinity of  
14 the cellulose within a reasonable time, improve the accessible area for the extraction of  
15 desirable components, and increase the hydrolysis rate after pretreatment (Agbor et al., 2011;  
16 Chin et al., 2020).

17 Organosolv pretreatment is a promising pretreatment for lignocellulosic materials. The  
18 concept of this process was derived from paper industry as an alternative to Kraft pulping and  
19 sulfite process, overcomes several inconveniences, such as severe water and air pollution  
20 (Bajpai, 2018; Tribot et al., 2019; Zhou et al., 2018). It is based on the extraction of lignin and  
21 hemicelluloses from wood materials using organic solvents with or without a catalyst (Zhang  
22 et al., 2016). However, the use of a catalyst (mineral acids, bases, or organic acids) allows to  
23 lower the pretreatment temperature (Tribot et al., 2019). The optimal temperature and  
24 exposure time are also interrelated. In general, a shorter exposure time requires a higher  
25 temperature and *vice versa* (Chin et al., 2020). Commonly used organic solvents in

1 Organosolv pretreatment are ethanol, methanol, acetone, or a mixture of water and organic  
2 solvent at temperatures ranging from 100 to 250 °C applied for a specific time (30–150 min).  
3 Low-boiling alcohols, such as methanol and ethanol, appear to be the most suitable organic  
4 liquids for use in Organosolv processes due to their low cost and easy recovery (Brosse et al.,  
5 2019; Zhao et al., 2009). Finally, ethanol is by far safer and preferred to methanol because of  
6 its low toxicity (Borand and Karaosmanoğlu, 2018; Zhao et al., 2009).

7 In the present work, a computer model able to simulate the conversion of second-generation  
8 sawdust feedstock to the bioethanol process is developed. Possible applications already  
9 documented refer to actual demonstration factories currently in operation or under  
10 construction in the European Union (Gnansounou and Dauriat, 2010). Such a tool is devoted  
11 to apprehending the main steps of the process, focusing on key aspects from the perspective  
12 of engineering thermodynamics. It provides a useful tool for the preliminary analysis of  
13 various plant configurations, allowing a potential reduction of costs for the plant. Mass and  
14 energy balances are modeled to allow a comparison between various technological solutions.  
15 This tool also notably makes it possible to assess the potential recovery of waste streams from  
16 the main process, in particular about lignin-rich residues. An Organosolv process based on  
17 ethanol/water mixtures as the solvent was selected to fractionate sawdust feedstock into its  
18 main components. Alio et al. (2019) investigated for the first time the fractionation of sawmill  
19 mixed feedstock of four softwood species using a microwave-assisted Organosolv  
20 pretreatment. Subsequently to this study, hydrolysis and fermentation steps of a pretreated  
21 feedstock were carried out (Abdou Alio et al., 2020). The ProSimPlus simulator is a powerful  
22 tool to carry out process simulation which is able to model the bioethanol production plant  
23 using lignocellulosic waste feed, such as sawdust or other residues. The chemical composition  
24 of sawdust and other streams of the process measured experimentally in previous works was  
25 used as the starting point in the simulation of the biorefinery to produce lignocellulosic

1 bioethanol using ProSimPlus as a process simulation software. The design of the plant  
2 included not only the Organosolv pretreatment, the enzymatic hydrolysis, and the  
3 fermentation steps, but also focused on distillation for ethanol recovery, water-saving, and  
4 energy integration. Lignin is recovered as a co-product for further use in added-value  
5 biocomposite materials (Tribot et al., 2019).

6 According to Rodrigues Gurgel da Silva et al. (2018), only a few researchers have turned their  
7 attention to investigate the synthesis, design, and process simulation of a large-scale  
8 Organosolv plant. Thus, the originality and the main objective of this paper is to fill this gap  
9 and develop a detailed process using the theoretical concepts and the experimental results  
10 (yields and compositions) from previous investigations (Abdou Alio et al., 2020; Alio et al.,  
11 2019) as a starting point to develop simulation of mass and energy balances based of an  
12 Organosolv pretreatment.

## 13 **2. Materials and methods**

14 The ethanol biorefinery process design in this study is adapted from previous works by NREL  
15 (National Renewable Energy Laboratory) according to Humbird et al. (2011) in order to  
16 include an Organosolv instead of a dilute-acid pretreatment. Thus all the data summarized in  
17 **Table 1** were collected from the technical investigation of the feedstock for all bioethanol  
18 processes step (Abdou Alio et al., 2020; Alio et al., 2019). As said earlier, these data were  
19 used as the starting point for the modeling and simulation activities in this assessment to  
20 obtain ethanol and other chemicals from the sawdust material.

21 As major assumptions made for this analysis, a typical large-scale production plant is  
22 considered: the amount of waste collected by the company is 200 tons/day; the moisture  
23 content of which is estimated at 40%, which gives 120 tons of dry biomass processed per day.

1 It is also considered an operating period of 350 days per year which is the equivalent of 8410  
2 hours/year (96% process uptime).

### 3 **2.1. Modeling and simulation**

4 The simulation of ethanol production from lignocellulosic biomass was carried out using  
5 ProSimPlus<sup>TM</sup>3.6. ProSimPlus is a French powerful software for running a multiphase process  
6 developed by *PROSIM S.A.* (Labège, France), including reliable thermodynamic data and  
7 comprehensive databases. For the current research, the NRTL (non-random two-liquid)  
8 property thermodynamic models were mainly used in the simulations. Ideal Gas was used for  
9 the simulation of combustion gases and NBS/NRC steam tables model was used in the  
10 simulation of water utilities. Components from ProSimPlus data banks were used whenever  
11 possible. The components and the properties which were not available in the software  
12 databases were obtained from the NREL data banks for production from lignocellulosic  
13 biomass and added in the model. Consequently, the BIODFMS3 physical properties database  
14 published by NREL was implemented and used in this study (Aden et al., 2002). As  
15 mentioned earlier, the simulations considered the setup of a plant processing 200 tons of  
16 feedstock per day.

### 17 **2.2. Process model overview**

18 The conventional NREL process model comprises nine following areas: feed handling,  
19 pretreatment and conditioning, enzymatic hydrolysis and fermentation, cellulase enzyme  
20 production, product recovery, wastewater treatment, storage, combustor, boiler and  
21 turbogenerator, and utilities.

22 In this study, only eight areas were studied, as shown in **Fig. 1**: commercial enzymes were  
23 used without local production. Simulation of water treatment is simplified to implement water



1 recycling. The process produces bioethanol and lignin, while the solid residues in addition to  
2 fresh bark to provide the hot utilities of the process; solvent and water are recycled. The  
3 process model description is summarized below.

#### 4 ***2.2.1. Part 0: feedstock handling***

5 The feedstock was collected and supplied from different surrounding sawmill industries as  
6 sawdust; hence, comminution was not included explicitly in this study. The transportation  
7 distance (between 50 and 100 km) of sawdust was estimated based on land patterns in France.

#### 8 ***2.2.2. Part 1: Organosolv pretreatment***

9 Sawdust mixture was stored in a sufficient amount to ensure the continuous operation of the  
10 biorefinery. The feedstock was transported by a conveyor to a mixer (**M101**) where an  
11 ethanol/water mixture (60:40 v/v) was added to the reactor in a solid-to-solvent ratio of 1:16  
12 (w/v) (**Fig. 2**). As a catalyst, sulfuric acid was added in a total of 0.25% (w/w) to dry wood  
13 sawdust matter. The mixture was sent to the pretreatment reactor (**R101**). The reactor  
14 operated at 175 °C and 14 bars of pressure with a total residence time of 60 min. Once the  
15 reaction is done, the pressure was reduced to atmospheric.

#### 16 ***2.2.3. Part 2: Lignin and solvent recovery***

17 The flashed reactor effluent was then separated in gas-liquid separator **S201** (**Fig. 3**). The  
18 evaporated solvent was sent to the solvent recovery columns and the pretreated mixture was  
19 filtered by passing through **S202** solid-liquid separator to obtain a solid-rich pulp (**PULP**),  
20 and another liquid-rich stream (**Liq.**) called black liquor. The wet solid fraction was mainly  
21 constituted of cellulose, while the liquid fraction contained dissolved hemicelluloses, lignin,  
22 and the remaining solvent. The solid stream was sent to a two-stage washing step (**M201** et  
23 **M202**), where a mixture of ethanol/water with the same concentration as in the reaction stage

1 was used, first, for cleaning the fibers and to recover some of the solvent and the remaining  
2 lignin. The resulting washing effluent was mixed with the liquid-rich stream, which can be  
3 treated to recover by-products (Organosolv lignin, furfural, HMF (5-hydroxyl-  
4 methylfurfural)), and solvent. The second stage of the washing process was performed with  
5 water in a solid/water ratio of 1:2 to remove the remaining solvent present in the solid-rich  
6 stream. After the washing process, the pulp was sent to the mixing tank reactor (**R301**) for  
7 enzymatic hydrolysis (**part 3**).

8 The total effluent streams composed of washing solvent, water, and a by-product as degraded  
9 cellulose (glucose), hemicellulose (xylose) and lignin were sent to the mixing tank (**M203**) so  
10 that all the by-products and solvent can further be recovered (**Fig. 3**). First, sulfuric acid was  
11 added to the mixing tank to allow the precipitation of lignin which will later be separated by  
12 filtration **S204**. This lignin is called Organosolv lignin. The liquid stream was treated with  
13 two distillation columns. The first distillation column (**C201**) was used to separate the solvent  
14 from the fermentable sugars (xylose and glucose) that were afterward sent to the fermentation  
15 reactor (**R302**). After, the first distillation, the sugars-free stream (**C018**) was sent to a second  
16 column (**C202**) where the ethanol at the top column was distilled to be recycled and re-used at  
17 the pretreatment step. The water stream (**C022**) at the bottom of the column was forwarded to  
18 the water treatment and recycling part (Part 6). This area allows reducing the fresh makeup  
19 solvent required.

#### 20 **2.2.4. Part 3: enzymatic hydrolysis and fermentation**

21 Several studies used separate hydrolysis and fermentation processes in their analysis. It was  
22 decided to use a similar setup in this study (**Fig. 4**), but data for modeling and simulation were  
23 modified, accounting for the raw materials from the Organosolv process applied to sawdust

1 and for the experimental results of fermentation and hydrolysis assays (Abdou Alio et al.,  
2 2020).

### 3 **2.2.4.1. Enzymatic hydrolysis**

4 The hydrolysis of the pulp stream can be performed by enzymes or acids (diluted or  
5 concentrated) catalysis. For bioethanol production using lignocellulosic materials, the  
6 enzymatic hydrolysis was demonstrated to be the most promising way for converting  
7 cellulose and hemicelluloses into simple sugars. Thus, the washed solid-rich was subjected to  
8 hydrolysis under a specific substrate charge. More specifically, the solid-rich stream reached  
9 the **R301** reactor (**Fig.4**) where it was diluted to a solid content of 10% (w/w) in a 50 mM  
10 buffer (pH 5). The enzyme used was Cellic® Ctec2 (A/S, Bagsvaerd, Denmark) with a total  
11 enzyme amount of 18.42 g per kg of the slurry. The enzymatic hydrolysis occurred at 50 °C in  
12 **R301** for 12 days. This step requires the use of several reactors in parallel for enzymatic  
13 hydrolysis to ensure continuous production.

### 14 **2.2.4.2. Fermentation**

15 Several microorganisms, such as bacteria, fungi, and yeast can ferment sugars to bioethanol.  
16 The most employed microorganism in fermenting carbohydrates from lignocellulosic is the  
17 yeast *Saccharomyces cerevisiae*, due to its high tolerance and robustness to inhibitory  
18 compounds. Consequently, after 12 days of hydrolysis, the stream from the enzymatic  
19 hydrolysis was further cooled up to 30 °C, mixed with the sugars recovered from C201, and  
20 sent to the fermentation reactors (**R302**), where *Saccharomyces cerevisiae* strain was added to  
21 the hydrolysate (**Fig. 4**). The fermentation took place in a 24 h time and had an  
22 initial inoculum level of 10% volume. Subsequently to the fermentation, the beer broth was  
23 sent to a beer well for separation and further ethanol purification.

### 2.2.5. Part 4: Ethanol recovery

After fermentation was finished, the fermentation broth consisted of ethanol, a variety of undesired co-products, and a large portion of water. Therefore, there are several technologies convenient for ethanol purification, such as pervaporation, multiple column distillation, liquid-liquid extraction, and so on, that use the differences in the physical and chemical properties of ethanol from unwanted products as the driving force for separation. Depending on the product that needs to be recovered and the composition of the mixture, an appropriate separation technology must be chosen. Hence, in this study, ethanol recovery was carried out in a multi-column separator (**Fig. 5**) similar to the one proposed by Aden et al. (2002). First, the fermentation broth was sent to a flash process to remove the main part of the carbon dioxide present in the flow (**C047**), and the resulting flow stream was sent to the mixer (**M401**). From this mixer, the broth was directed to the first distillation column (**C401**) where three streams were separated. The overhead stream was rich in carbon dioxide and contained about 13% (w/w) ethanol. This flow could be mixed with the fermentation vent and sent to a vent scrubber (**C402**), where water was added in a counter-current configuration to recover ethanol present in the vapor phase. The effluent from the scrubber was returned to the beer well. The bottom product of the distillation column contained the unreacted and dissolved solids, which can be dried and burnt for energy production (Part 6). Finally, an intermediate withdrawal containing 42% (w/w) ethanol, i.e. 99.5% of the total ethanol from the **R302** effluent, was sent to the rectification column (**C403**). It was then purified to the nearest azeotropic point, and then, sent to the molecular sieves to decrease residual water. Two streams left the molecular sieves: ethanol at 99.5% (w/w), and lower concentration ethanol (72%) that could be returned to the distillation column. The rectification column bottom containing less than 0.1% (w/w) ethanol was sent to water recycling.

### 1 **2.2.6. Parts 5, 6 and 7**

2 The residual solids and dissolved sugars from the beer bottom were concentrated through a  
3 triple-effect evaporator as proposed by Aden et al. (2002). Solids were removed after the first  
4 effect and concentrated syrup was recovered after the third effect; both were mixed and sent  
5 to a combustion chamber at a moisture content of 51%.

6 Part 6 was dedicated to water treatment. Main assumptions are total removal of organic  
7 dissolved matter and 1.0% of entering water is not recycled. The current publication provides  
8 a supplementary figure that shows the generalized flowsheet (**supplementary data 1**)

9 Finally, part 7 provided the hot utilities of the process by burning the unreacted solids and  
10 sugars concentrated in Part 5. Another wood waste fresh bark was also burnt in the  
11 combustion chamber to meet energy requirements. Additional 200 t/d of fresh bark, whose  
12 composition is given in **Table 1**, were necessary to produce 900 t/d of steam at 60 bars and  
13 478 °C. This steam was used in a Rankine cycle at four levels of pressure respectively 60, 10,  
14 2, and 0.125 bars. The cycle was simulated with three turbines to produce electricity and two  
15 withdrawals at intermediate pressures of 10 and 2 bars, respectively, to provide hot utilities to  
16 the process (**supplementary data 2**).

### 17 **2.3. Key technical parameters**

18 The key technical parameters used in ProSimPlus simulations were obtained from Alio et al.  
19 (2020) and Alio et al. (2019), as mentioned earlier. In the pretreatment stage, most of the  
20 hemicelluloses are degraded into xylose and furfural, and the crystalline structure of most of  
21 the cellulose is broken down, increasing enzyme accessibility to the cellulose for hydrolysis  
22 step before the fermentation step. Enzymatic hydrolysis includes cellulose and hemicellulose  
23 conversion into sugars. Fermentation of C5 and C6 sugars are also considered. **Table 2**  
24 summarizes the data needed to simulate the process.

## 2.4. Heat integration

The ProSimPlus simulation showed that hot and cold external utilities were required to meet the needs of ethanol production utilities. Since external utilities are expensive and require large quantities, the ability to integrate heat between processes was investigated using process pinch technology. In this article, an integrated ProSimPlus module was used to apply process pinch analysis. Pinch Analysis established a hot and cold composite curve from the available hot and cold streams in the flow diagram. Later, the two composite curves built from the hot and cold streams were compared with each other at a minimum approach temperature to determine the possible amount of heat exchange (integration) between the hot and cold flows. This integration of heat between processes reduced the external hot and cold energy requirements of a production circuit. In this work, the minimum approach temperature  $\Delta T_{\min}$  of 10 °C was used for the pinch analysis. Hot effluents of the process were integrated in the calculations by cooling down to a temperature of 25 °C. Phase change was linearized for the calculations in the pinch module.

## 3. Results and discussion

This process model was set up to assess the viability of a second-generation ethanol production process, which uses a mixture of sawmill waste of softwood as the feedstock, biomass widely available in Auvergne-Rhône-Alpes region (France) as a forest residue or from dedicated short-rotation forestry. An efficient conversion process of the cellulosic and hemicellulosic biomass into liquid ethanol, combined with a valorization of the lignin content, would allow an energy conversion that would meet the ever-growing energy demand. The simulation considered the scale-up of the laboratory and pilot-scale systems used to estimate the data to produce significant results in terms of mass and energy balances for the design and

1 the development of new industrial perspectives. However, the results presented in this section  
2 will focus first on mass balances.

### 3 **3.1. Ethanol production mass balance**

4 The analysis of the ethanol production simulation process from lignocellulosic residues is  
5 turned towards process productivity. In this case, from the initial 200 tons of sawdust  
6 feedstock per day, simulations showed that the product stream from Organosolv pretreatment  
7 yielded 33.11 tons of ethanol per day (1380.2 kg of ethanol/h), as shown in the **C063** stream  
8 (**Table 3**). The ratio between ethanol production expressed in liters and the mass of dry  
9 biomass consumed expressed in tons is, therefore, 349 L/ton. In detail, almost all the glucose  
10 was consumed during the fermentation. This was mainly obtained in the enzymatic hydrolysis  
11 step. In the pretreatment step, only a small fraction of cellulose was converted into glucose. In  
12 this sense, the cellulose conversion was limited to 16.2%. In the enzymatic hydrolysis, the  
13 conversion of cellulose into glucose reached 80%. However, it arises that 35% of the  
14 hemicellulose of the raw materials remained unconverted in the process, probably due to the  
15 lack of efficiency of *S. cerevisiae* for converting xylose into ethanol. It must be mentioned  
16 that the combination of columns **C201** and **C202** allowed to remove the furfural from the  
17 sugars produced in the pretreatment reactor (0.7 t/d of furfural in stream C011 and 0 in stream  
18 C046). Consequently, the conversion into ethanol could be raised by recycling those sugars  
19 (stream C044 and C046) into the fermenter **R303** without any risks of inhibition.

20 The results thus highlighted an ethanol production of approximately 11,600 tons of  
21 ethanol/year with a biomass feed of 42,052 tons of dry biomass per year, which is in the  
22 common range found for this kind of process. For example, for the same quantity of dry  
23 feedstock, Porzio et al. (2012) obtained less than 10,000 tons of ethanol/year by using a two-  
24 stage dilute acid steam explosion pretreatment and a productivity of only 303–316 L/t dry

1 biomass. The results also outperform the simulations of the diluted acid process reported by  
2 Rodrigues Gurgel da Silva et al. (2018). Conversely, based on an Organosolv process  
3 modeling on the same feedstock quantity, their simulations yielded a higher ethanol  
4 production of 11,922 tons of ethanol/year. But this ethanol amount was achieved because the  
5 authors considered a high conversion of both xylan and cellulose into convertible sugars, thus  
6 into ethanol. First, it must be pointed out that the present work uses a mixture of softwood  
7 species as the feedstock, contrary to the literature which always deals with a unique wood  
8 specie. This drawback was partially counterbalanced by a higher ethanol concentration  
9 achieved in the fermentation in the present study (12.7 % w/w) than in Rodrigues Gurgel da  
10 Silva et al. (2018)'s case (11.1% w/w), which highlights the effectiveness of our pretreatment.  
11 In addition, C5 fermentation was not favored in this work, as stated above, so that ethanol  
12 production could still be enhanced by fermenting xylose in the residue (C057) so that ethanol  
13 productivity could outperform the literature in this case.

14 Finally, **Table 4** illustrates these statements by displaying a detailed comparison of ethanol  
15 concentration in the fermenter, and ethanol productivity values with the literature.

### 16 **3.2. Water mass balance**

17 Water consumption in the biochemical conversion process is generally considered high  
18 compared to a thermochemical pathway process or even a petroleum-based fuel process, in  
19 terms of a liter of water used per liter of fuel produced (L/L).

20 The overall water balance according to the simulation results indicate the water consumption.  
21 Additional water enters the process via the raw material and raw chemicals, both as free  
22 water, and "potential" water, i.e. the burning product of lignin and unconverted sugars. Also,  
23 water is utilized in the hydrolysis reactions. Based on actual make-up water flow, the current  
24 design needed water consumption of 8417 kg/h versus 8760 kg/h from Humbird et al. (2011)  
25 for a plant that has the same feedstock throughput, using an acid-based pretreatment.



1 Conversely, 8332 kg of water/h were consumed according to Rodrigues Gurgel da Silva et al.  
2 (2018) flowsheet design using an Organosolv pretreatment under the same feedstock  
3 throughput. However, based on the detailed ethanol yield from the ProSimPlus simulation, the  
4 water consumption in the design, about 4.8 liters of water used per liter of ethanol, was nearly  
5 the same as that obtained in the Organosolv simulations of the literature and far lower than in  
6 the NREL study (7.65 L/L) or with diluted acid pretreatments (**Table 4**). The key advantage  
7 of the Organosolv pretreatment, as already emphasized by the literature, is that it saves water  
8 in comparison to conventional acid or steam explosion processes. In order to be economically  
9 feasible, the overall process needed to recover as much as possible the solvent (water and  
10 ethanol) used in the process. Based on the simulation data gathered in **Table 3**, up to 99.9% of  
11 the ethanol and 99% of the water that were used initially were recycled. But the recovery  
12 increased the whole process energy demand.

### 13 **3.3. Energy balance**

14 All hot and cold process streams extracted from a flowsheet model of the studied case design  
15 were listed in **Table 5**. By summing all thermal loads of all cold and hot flows, the maximum  
16 hot and cold utility requirements of the studied case were obtained. Thus, the hot and cold  
17 utilities demand represented 19.1 and 18.7 MW, respectively. Among this demand, distillation  
18 columns were the most important part of the energy consumption respectively 65% for boilers  
19 and 71% for condensers. The energy per ethanol produced was 27.4 kWh per kg of ethanol.

20 This energy demand is consistent with the data from the literature (Table 4). For an  
21 Organosolv process, Rodrigues Gurgel da Silva et al. (2018) found 18.7 and 15.0 MW as  
22 utility requirements and 23.9 kWh per kg of ethanol. The difference between their simulations  
23 and this work is the simulation of part 5 which is the evaporation of the solids and syrup that  
24 requires hot and cold utilities. Without part 5, the energy falls to 24.0 kWh/kg of ethanol. On

1 the other hand, the acid diluted process is much less energy-intensive (13.2 kWh/kg of  
2 ethanol). The origin of the rather high energy demand of the whole process can be identified  
3 in the solvent recycling unit. This counterbalances the higher ethanol productivity and water  
4 efficiency of the Organosolv process. In particular, ethanol as the solvent must be recycled  
5 and an important amount of water is required in the pretreatment process to dilute ethanol and  
6 to cause lignin precipitation. The water terminates in the solvent recycling system, which  
7 requires the highest energy input.

### 8 ***3.4. Pinch Analysis and further heat integration***

9 Based on the data of **Table 5**, the next step in the Pinch Analysis is to determine energy  
10 targets. The energy targets represent the minimum amount of utilities required to meet the  
11 process flow requirements (Ghannadzadeh and Sadeqzadeh, 2017). As mentioned in **section**  
12 **2.4**, this was done by plotting composite cold (blue) and hot (red) process curves, as shown in  
13 **Fig.6a**.

14 The hot and cold composite curves give an idea of the heat availability and heat demand in the  
15 process, respectively. The pinch temperature was evaluated at 90.2 °C. According to **Fig. 6a**,  
16 the potential of heat recovery is not very high on this process as the overlay between hot and  
17 cold composite curves is rather partial. Maximum energy recovery (MER) was estimated at 2.7  
18 MW. Consequently, the hot and cold utilities demand could be reduced from 19.1 and 18.7 MW  
19 to respectively 16.4 and 16.0 MW which would represent less than 15.0 % of heat integration  
20 on the process. There are two main reasons for this poor heat recovery potential:

21 *(i)* the flash after the pretreatment reactor (**R101**) does not allow to use a hot stream at 175 °C  
22 that is why the hot composite curve for **Fig. 6a** does not reach 175 °C that would raise the MER.  
23 The evaporated solvent after the valve **V101** represents nonetheless a heat integration because  
24 it reduces the energy demand of column **C201** by vaporizing the feed of column.

1 (ii) the level of temperature (pressure range) on the distillation columns (C201, C202, C401,  
2 and C403) does not allow the heat transfer between condensers and boilers although enthalpy  
3 fluxes of condensers are higher than the boilers' one.

4 The grand composite curve confirms that heat recovery in the process is limited. Only small  
5 pockets of self-sufficient process to process heat transfer can be seen in Fig. 6b in the cooling  
6 area: between 90 and 86 °C, between 73 and 57 °C for example. The residual cooling necessity  
7 (15.9 MW) was obtained thanks to water from 12 to 25 °C with a volumetric flow rate of 25375  
8 m<sup>3</sup>/d. Concerning the hot utilities, two levels of pressure steam were selected: 10 bars high  
9 pressure (HP) steam at a condensing temperature of 179.9 °C for temperatures higher than 106.9  
10 °C and 2 bars low pressure (LP) steam at a condensing temperature of 120.2 °C for temperatures  
11 between 106.9 °C and the pinch temperature. The unreacted sugars and solids could only  
12 partially generate required steam that is why available fresh bark was added to match hot energy  
13 requirements. The HP and LP hot utilities steam represented in Fig. 6b were those produced by  
14 the Rankine cycle of part 7. 175 t/d and 470 t/d of HP and LP steam, respectively, were  
15 withdrawn for heat transfer on the process. The condensed vapors were then used to preheat  
16 water before the cycle boiler. The Rankine cycle could also produce 6887 kW of electricity, but  
17 10265 m<sup>3</sup>/d of freshwater was also needed for the cycle condenser.

### 18 ***3.5. Effect of co-product markets in production cost***

19 According to Table 3, it can be noticed that the amount of unreacted Residue in the  
20 production stream C057 of 21 tons/day which corresponds to 7,344 tons/year is mainly  
21 composed of 58% lignin, 32% cellulose, 8% hemicellulose, and 1.7% others. In the present  
22 simulations, these unreacted solids were further burned in a perspective of cogeneration,  
23 increasing the capacity of heat integration for the process. In another perspective, this solid  
24 residue can be used as pellets, but this would increase external energy demand.

1 Furthermore, the potentiality for selling by-products, which are transformed into value-added  
2 by-products in the ethanol production plant, also has the prospective for reducing production  
3 costs. Outstandingly, expenses occurred in treating industrial effluents to avoid serious  
4 pollution problems. For example, Organosolv lignin (18.96 tons/day which represents 6,644  
5 tons of lignin/year) can be purified and transformed into marketable products, i.e. powder  
6 lignin that helps balance the economics of the overall production process. Potential  
7 applications have been reviewed by Tribot et al. (2019). This will result in cost savings that  
8 come either from reduced waste handling costs or from the generation of revenue from the  
9 sale of by-products. The consequence is that the economic analysis of the Organosolv process  
10 is still difficult to assess, as it strongly depends on the co-products.

#### 11 **4. Conclusion**

12 Process simulations demonstrate that an ethanol/water Organosolv pretreatment could be  
13 successfully applied to a mixture of softwood species in a forest biorefinery. An ethanol  
14 productivity of 349 L/ton of dry biomass is achieved. The biorefinery design enhances water  
15 and ethanol savings (4.8 L/L ethanol, and 99% recovery, respectively), at the expense of  
16 energy requirements (10.9 and 8.6 kWh/L ethanol for hot and cold utilities, respectively). The  
17 plant converts 70,088 tons/year biomass into 11,400 tons/year ethanol. A better understanding  
18 of the process was achieved, but further research on the valorization of co-products must help  
19 balance the economics of the overall process.

#### 20 **Acknowledgments**

21 This work was financed by Auvergne Rhône-Alpes Regional Council, and the European  
22 Regional Development Fund (FEDER/ERDF) to promote the valorization of local feedstocks  
23 (wood-wastes) on a regional scale.

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1 **Figure captions**

2 **Fig. 1:** Simplified bioethanol production hierarchy block diagram

3 **Fig. 2:** Simulation flowsheet of the Organosolv pretreatment

4 **Fig. 3:** Simulation flowsheet for lignin and solvent recovery

5 **Fig. 4:** Simulation flowsheet for hydrolysis and fermentation steps

6 **Fig. 5:** Simulation flowsheet of the setup for ethanol separation and purification

7 **Fig. 6:** Pinch Analysis results curves of the biorefinery: a) composite curves representing hot and  
8 cold streams with  $\Delta T_{\min}=10$  °C; b) grand composite curve with hot and cold utilities simulated  
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1 **Table captions**

2 **Table 1:** Main process data for simulation of ethanol production from wood waste

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5 **Table 2:** Key technical parameters

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9 **Table 3:** Process streams mass balance in tons/d of the flowsheet

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12 **Table 4:** Comparison of process data between the present studied case and one from the  
13 literature (Rodrigues Gurgel da Silva et al., 2018) for an alternative Organosolv and a diluted  
14 acid pretreatment

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20 **Table 3:** Hot and cold streams data of the process including all the different process  
21 sections (supply temperature  $T_{in}$ ; target temperature  $T_{out}$ ; heat energy  $Q$ ; heat capacity  $F \cdot C_p$ )

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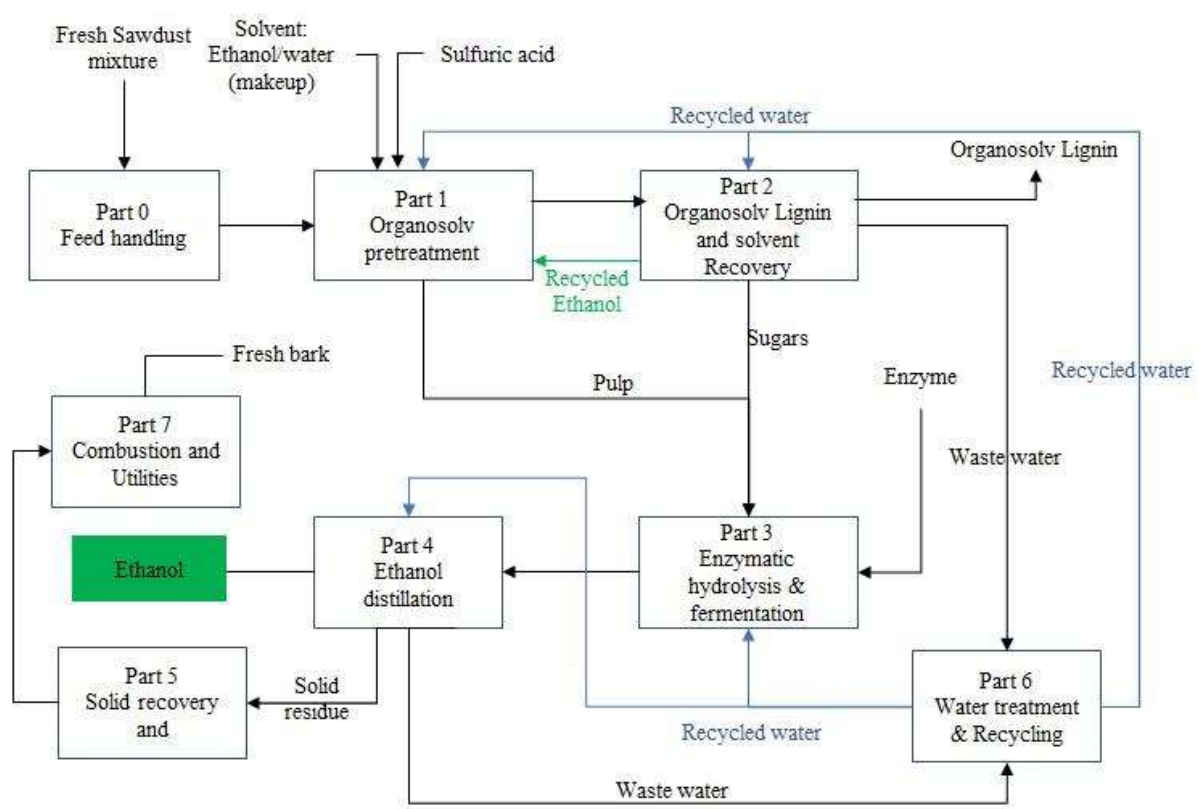
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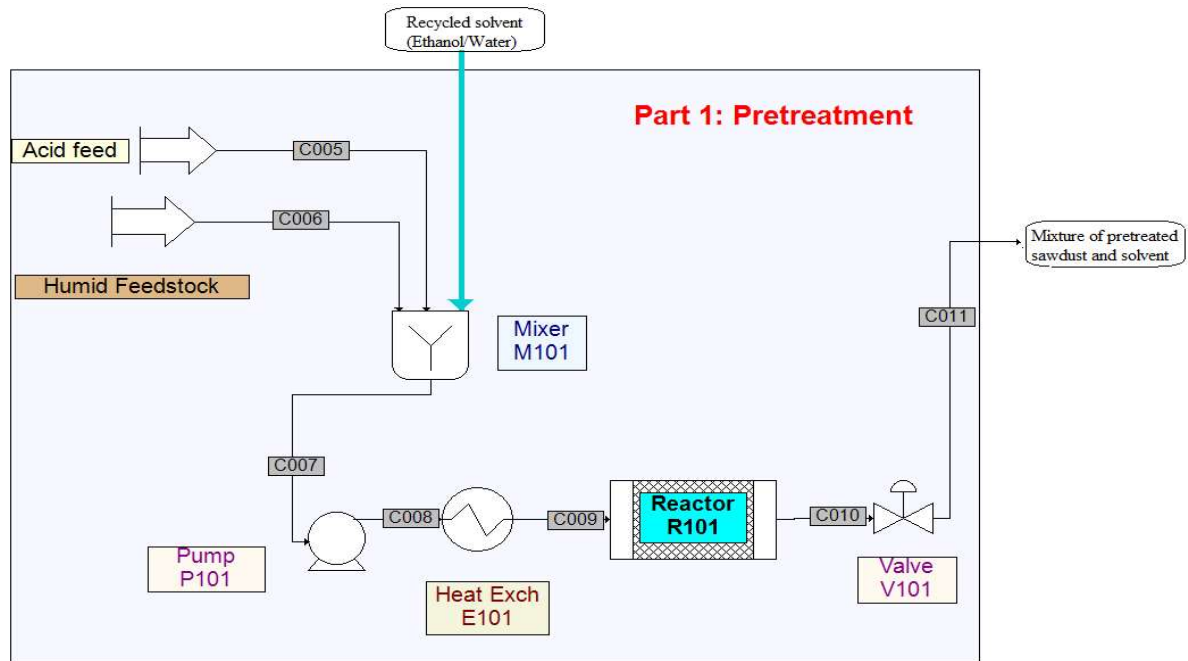
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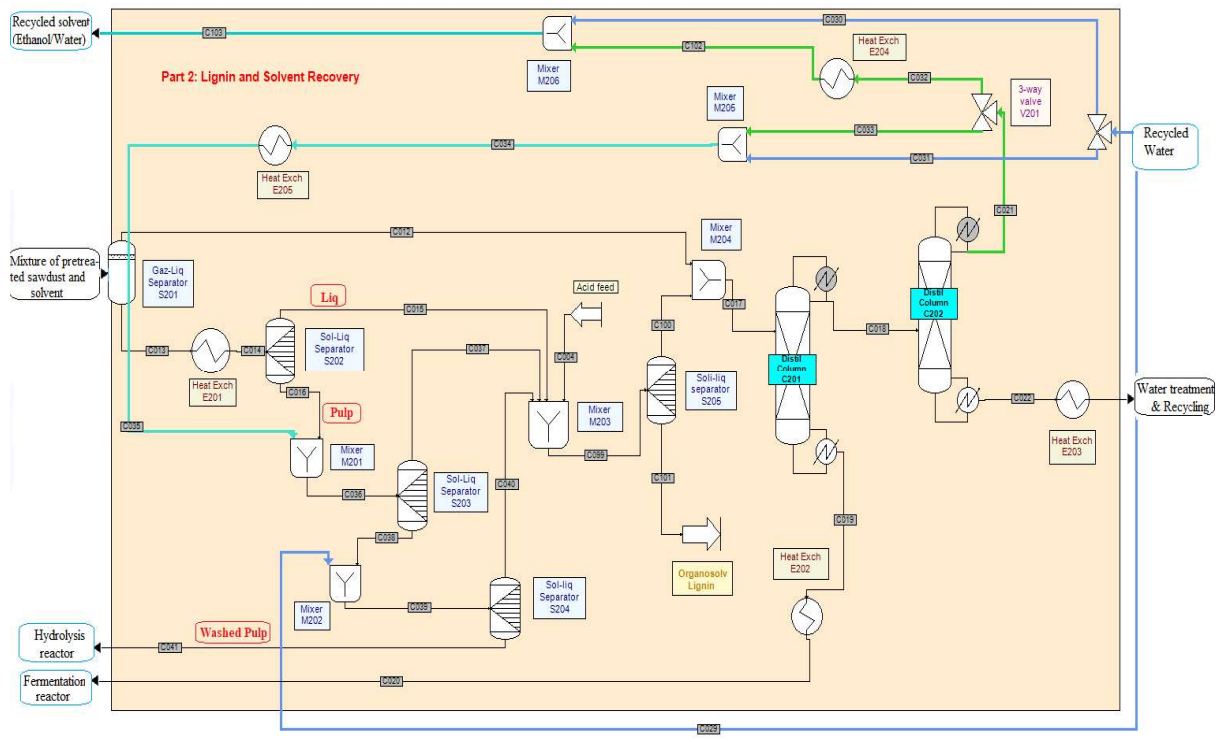
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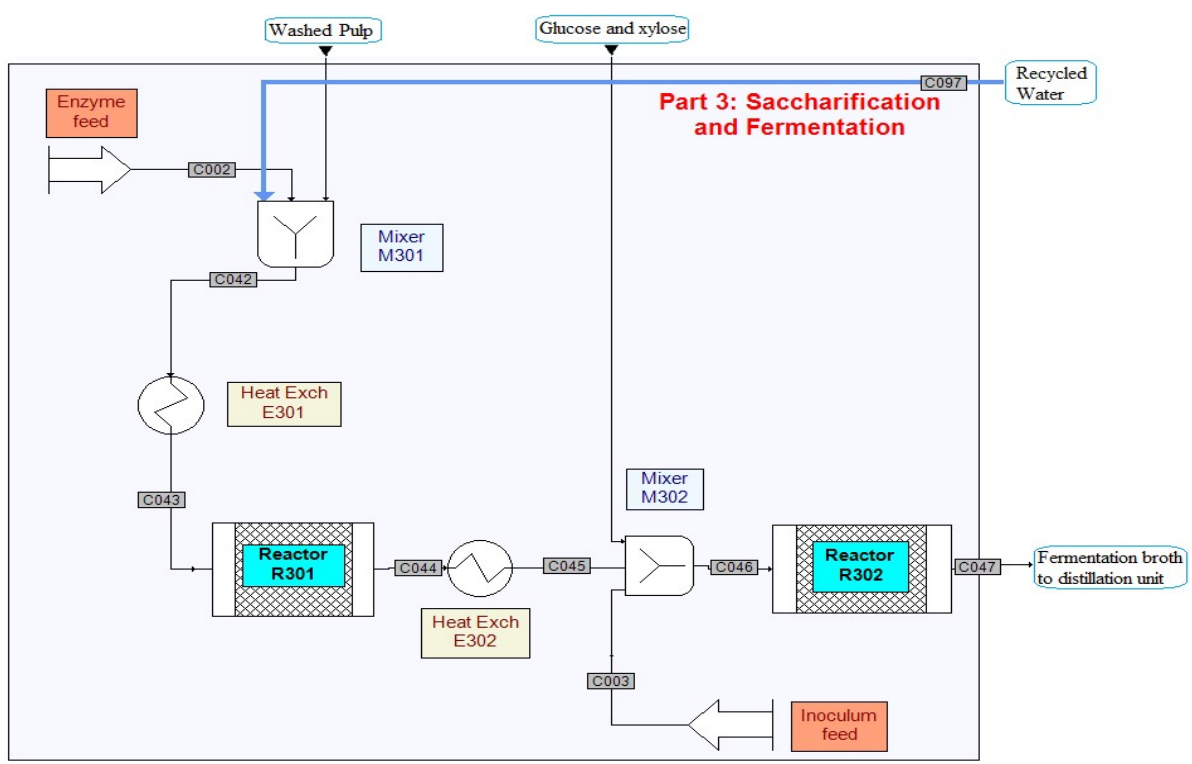
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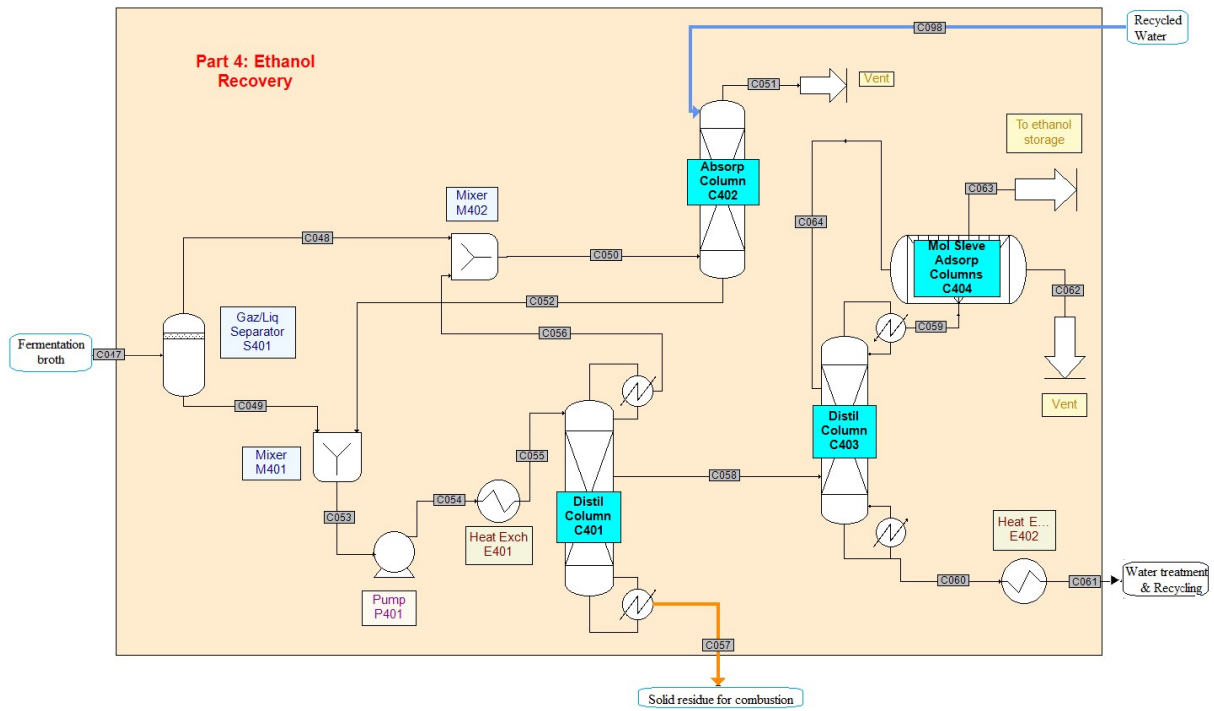
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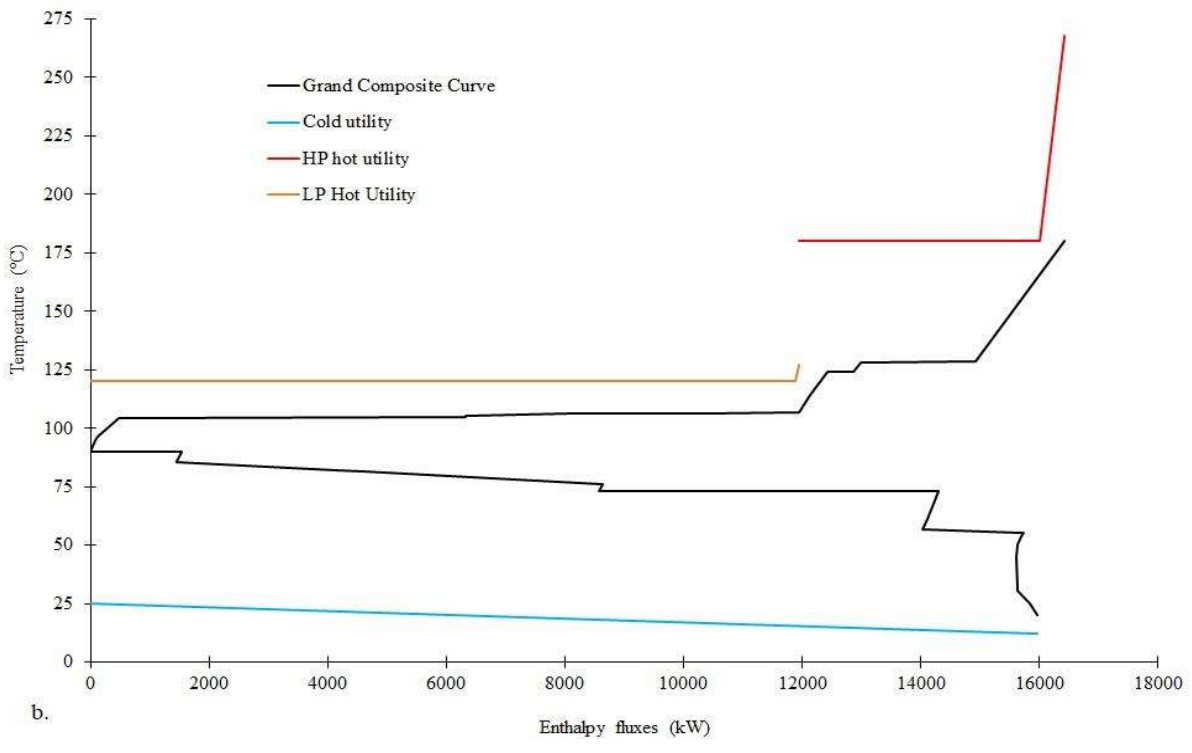
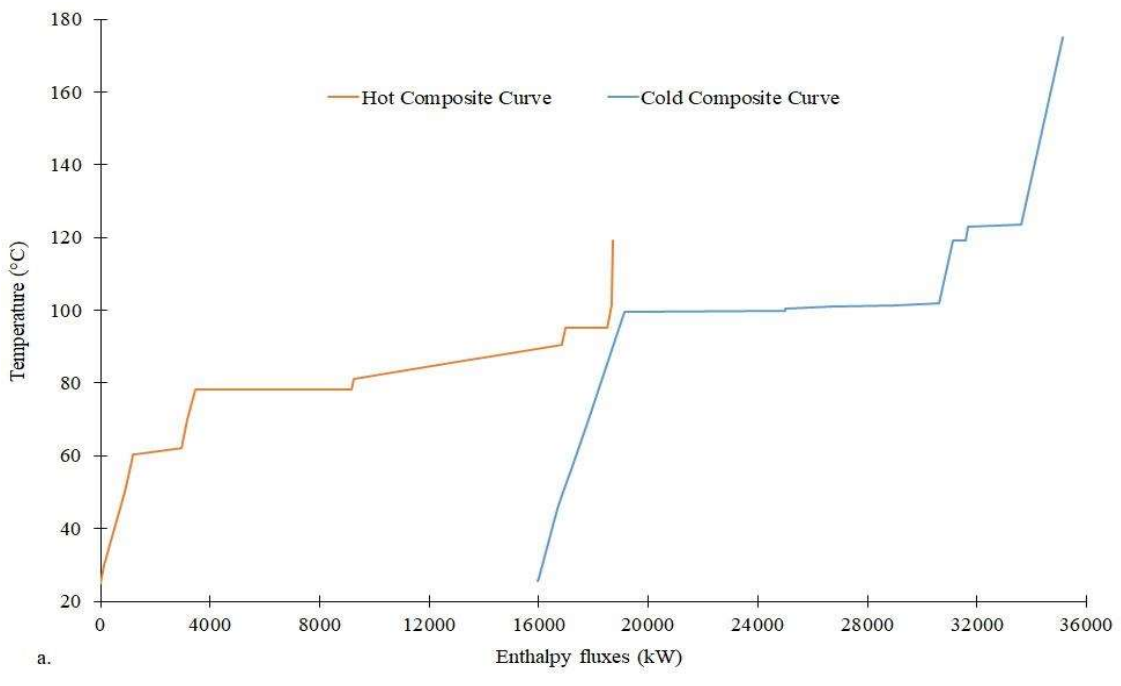




Table 1

| Features                   | Values   |                                |   |
|----------------------------|--|--------------------------------|---|
| <b><u>Feedstock:</u></b>   |  |                                |   |
| <b>Sawdust</b>             | Cellulose 44.3% ± 0.5% (dry wt.), hemicellulose 25.6% ± 0.2% (dry wt.),  |                                |   |
| Composition                | lignin 26% ± 3% (dry wt.)  |                                |   |
| Moisture content           | 40%  |                                |   |
| <b>Bark</b>                | Cellulose 28.3% ± 1.2 % (dry wt.), hemicellulose 13.0% ± 0.8% (dry wt.), |                                |   |
| Composition                | lignin 52% ± 4.2% (dry wt.)  |                                |   |
| Moisture content           | 20%  |                                |   |
| <b><u>Reactors:</u></b>    |  |                                |   |
| Type                       | <b>Pretreatment</b>  | <b>Enzymatic</b>               | <b>Ethanol by</b>                         |
|                            | <b>Organosolv</b>  | <b>Hydrolysis</b>              | <b>Fermentation</b>                       |
| Solvent                    | Ethanol/water<br>(60:40)   | Water                          | Water                                     |
| Catalyst                   | H <sub>2</sub> SO <sub>4</sub>   | Cellulase and<br>hemicellulase | <i>Saccharomyces</i><br><i>cerevisiae</i> |
| Concentration              | 0.25%  | 18.42 g<br>per kg of slurry    | 10% of fermentation<br>volume             |
| Temperature                | 175 °C   | 50 °C                          | 30 °C                                     |
| Pressure                   | 13.8 atm   | 1 atm                          | 1 atm                                     |
| Residence time             | 60 min   | 12 days                        | 24 h                                      |
| pH                         | -  | 5                              | 5   |
| <b><u>Separations:</u></b> |  |                                |   |
| <b>Solvent recovery</b>    | <b>Concentration C201</b>  |                                | <b>Rectification C202</b>                 |
| Number of stages           | 15   |                                | 40  |
| Stage efficiency           | 57%  |                                | 57%                                       |
| Feed stage                 | 2  |                                | 25  |
| Reflux ratio               | 0.03   |                                | 1.0                                       |
| Condenser                  | Total  |                                | Total                                     |
| Pressure                   | 1.0 atm  |                                | 1.0 atm                                   |

| <b>Ethanol recovery</b>    | <b>Concentration C401</b>  | <b>Rectification C403</b> |
|----------------------------|--|---------------------------|
| Number of stages           | 32   | 60                        |
| Stage efficiency           | 48%  | 57%                       |
| Feed stage                 | 2  | 40                        |
| Reflux ratio               | 3.0  | 3.2                       |
| Condenser                  | Partial  | Partial                   |
| Pressure                   | 2.1 atm  | 1.9 atm                   |
| Additional                 | Withdrawal stage 8   | Feed stage 20             |
| <b>Scrubber</b>            | <b>Absorption column C402 (5 stages, 1.0 atm, non-isothermal)</b>          |                           |
| <b>Ethanol dehydration</b> | <b>Pressure swing adsorption molecular sieves C404 (95.0 °C, 1.53 atm)</b> |                           |

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**Table 1:**

| Technical parameters        |                         | Yield |
|-----------------------------|-------------------------|-------|
| <b>Pretreatment</b>         | Cellulose to glucose    | 16.5% |
|                             | Hemicellulose to xylose | 81%   |
|                             | Lignin removal          | 45.8% |
|                             | Ash                     | 0%    |
|                             | Extractives             | 100%  |
| <b>Enzymatic hydrolysis</b> | Cellulose to glucose    | 80%   |
|                             | Hemicellulose to xylose | 60%   |
| <b>Fermentation</b>         | Glucose to ethanol      | 99%   |
|                             | Xylose to ethanol       | 64%   |

Table 3

| Streams                | C006             | C009  | C011  | C016 | C015  | C101           | C020 | C044  | C046  | C047  | C055  | C057    | C058  | C063    | C024         |
|------------------------|------------------|-------|-------|------|-------|----------------|------|-------|-------|-------|-------|---------|-------|---------|--------------|
|                        | Fresh<br>sawdust | E101  | R101  | Pulp | Liq   | Org.<br>lignin | E202 | R301  | M302  | R302  | C401  | Residue | C403  | Ethanol | Water Treat. |
| Water                  | 76.7             | 139.5 | 136.0 | -    | 169.7 | -              | 56.7 | 107.7 | 232.1 | 232.1 | 274.1 | 224.5   | 49.6  | 0.2     | 157.7        |
| Carbon dioxide         | -                | -     | -     | -    | -     | -              | -    | -     | -     | 31.6  | 0.5   | -       | 0.0   | -       | -            |
| Ethanol                | -                | 180.0 | 180.0 | -    | 74.1  | -              | -    | -     | -     | 33.1  | 33.2  | 0.0     | 33.1  | 33.1    | -            |
| Sulfuric acid          | -                | 0.5   | 0.5   | -    | 0.7   | 0.7            | 0.0  | -     | 0.0   | 0.0   | 0.0   | 0.0     | -     | -       | -            |
| Glucose                | -                | -     | 8.6   | -    | 8.6   | -              | 8.6  | 41.4  | 50.0  | 0.5   | 0.5   | 0.5     | -     | -       | -            |
| Xylose                 | -                | -     | 26.2  | -    | 26.2  | -              | 26.2 | 5.8   | 31.9  | 11.2  | 11.2  | 11.2    | 0.0   | -       | -            |
| Cellulose              | 53.2             | 53.2  | 44.4  | 44.4 | -     | -              | 0.0  | 6.7   | 6.7   | 6.7   | 6.7   | 6.7     | 0.0   | -       | -            |
| Hemicellulose          | 30.7             | 30.7  | 6.8   | 6.8  | -     | -              | -    | 1.7   | 1.7   | 1.7   | 1.7   | 1.7     | -     | -       | -            |
| Lignin                 | 31.1             | 31.1  | 31.1  | 12.2 | 18.9  | 18.9           | 0.0  | 12.2  | 12.2  | 12.2  | 12.2  | 12.2    | 0.0   | -       | -            |
| Furfural               | -                | 0.0   | 0.7   | 0.0  | 0.4   | -              | 0.0  | 0.0   | 0.0   | 0.0   | 0.0   | -       | 0.0   | -       | -            |
| Cellulobiose           | -                | -     | -     | -    | -     | -              | -    | 0.5   | 0.5   | 0.5   | 0.5   | 0.5     | -     | -       | -            |
| Enzyme                 | -                | -     | -     | -    | -     | -              | -    | 1.2   | 1.2   | 1.2   | 1.2   | 1.2     | 0.0   | -       | -            |
| <i>S. cerevisiae</i>   | -                | -     | -     | -    | -     | -              | -    | -     | 0.4   | 0.4   | 0.4   | 0.4     | -     | -       | -            |
| Acetic acid            | -                | -     | -     | -    | -     | -              | -    | -     | -     | 5.5   | 5.5   | 5.0     | 0.5   | -       | -            |
| HMF                    | -                | -     | 0.8   | 0.8  | -     | -              | 0.0  | 0.8   | 0.8   | 0.8   | 0.8   | 0.8     | 0.0   | -       | -            |
| Total flow (t/d)       | 191.7            | 435.0 | 435.0 | 64.1 | 298.6 | 19.6           | 91.4 | 177.8 | 339.9 | 339.9 | 350.8 | 267.0   | 83.2  | 33.3    | 157.7        |
| T (°C)                 | 25.0             | 175.0 | 174.6 | 33.2 | 48.5  | 48.5           | 30.0 | 50.0  | 30.0  | 45.3  | 100.0 | 123.4   | 112.1 | 92.1    | 25.0         |
| P (atm)                | 1.0              | 13.8  | 13.8  | 1.0  | 1.0   | 1.0            | 1.0  | 1.0   | 1.0   | 1.0   | 2.1   | 2.1     | 1.9   | 1.7     | 1.0          |
| Vap. fraction<br>(w/w) | -                | 0.4   | 0.4   | -    | -     | -              | -    | -     | -     | 0.1   | 0.0   | -       | 1.0   | -       | -            |



Table 4

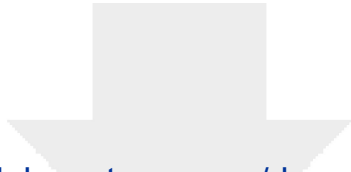
|                                 | Studied case <sup>a</sup> | Organosolv <sup>b</sup> | Diluted acid <sup>b</sup> |
|---------------------------------|---------------------------|-------------------------|---------------------------|
| Ethanol productivity (kg/h)     | 1380.2                    | 1417.2                  | 1271.7                    |
| Ethanol concentration (% w/w)   | 12.7                      | 11.1                    | 5.4                       |
| Hot utilities (MW)              | 19.1                      | 18.7                    | 11.7                      |
| Cold utilities (MW)             | 18.7                      | 15.0                    | 5.1                       |
| Total energy (MW)               | 37.8                      | 33.7                    | 16.8                      |
| Energy (kWh)/kg of ethanol      | 27.4                      | 23.9                    | 13.2                      |
| Water consumption (kg/h)        | 8417.0                    | 8332.2                  | 22090                     |
| Ethanol/water ratio (L/L)       | 4.8                       | 4.7                     | 19.0                      |
| Ethanol/dry biomass ratio (L/t) | 349                       | 358                     | 232                       |

<sup>a</sup> Results obtained for 120 tons of dry biomass per day of sawmill mixed feedstock

<sup>b</sup> Results obtained from 2124 tons of dry biomass of spruce feedstock per day have been normalized based on 120 tons of dry biomass per day

Table 5

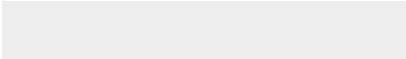
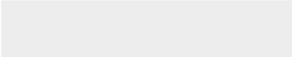
| Streams           | Physical status | $F \cdot C_p$ (kW/ °C) | $T_{in}$ (°C) | $T_{out}$ (°C) | Q (kW)   |
|-------------------|-----------------|------------------------|---------------|----------------|----------|
| Bo. Column C201   | Liq             | 2995.0                 | 100.5         | 102.0          | 4236.7   |
| Bo. Column C202   | Liq             | 14 321.5               | 99.5          | 99.9           | 5831.3   |
| C034              | Liq             | 2.9                    | 56.4          | 70.0           | 39.7     |
| C008              | Liq             | 29.1                   | 25.7          | 175.0          | 4341.5   |
| Bo. Column C403   | Liq             | 8 701.1                | 119.0         | 119.1          | 438.0    |
| C042              | Liq             | 6.4                    | 26.3          | 50.0           | 151.0    |
| C054              | Liq             | 15.3                   | 45.5          | 100.0          | 833.7    |
| Bo. Column C401   | Liq             | 4 836.4                | 123.0         | 123.4          | 1925.4   |
| C065              | Liq-Vap         | 4 327.6                | 101.1         | 101.4          | 1340.0   |
| Cond. Column C201 | Liq-Vap         | 768.50                 | 90.6          | 81.0           | - 7323.9 |
| Cond. Column C202 | Liq-Vap         | 55 146.0               | 78.4          | 78.2           | - 5710.1 |
| C022              | Liq             | 7.7                    | 99.9          | 25.0           | - 579.4  |
| C032              | Liq             | 6.6                    | 78.2          | 25.0           | - 351.1  |
| C013              | Liq             | 6.1                    | 90.8          | 70.0           | - 126.8  |
| C101              | Liq             | 0.3                    | 58.8          | 25.0           | - 9.8    |
| C019              | Liq             | 3.3                    | 101.9         | 30.0           | - 237.9  |
| Cond. Column C403 | Liq-Vap         | 224 760.0              | 95.2          | 95.2           | - 1537.9 |
| C063              | Liq             | 1.1                    | 95.2          | 25.0           | - 75.9   |
| C060              | Liq             | 2.4                    | 119.10        | 25.0           | - 227.1  |
| C044              | Liq             | 6.6                    | 50.0          | 30.0           | -131.9   |
| Cond. Column C401 | Liq-Vap         | 0.5                    | 102.7         | 60.4           | - 20.0   |
| C068              | Liq             | 8.8                    | 101.4         | 87.0           | - 126.0  |
| C074              | Liq             | 3.0                    | 100.1         | 25.0           | - 223.6  |
| C083              | Vap             | 1096.7                 | 62.00         | 60.4           | - 1719.5 |
| C090              | Liq             | 3.1                    | 60.5          | 25.0           | - 109.0  |
| C081              | Liq             | 3.0                    | 86.4          | 25.0           | - 180.0  |



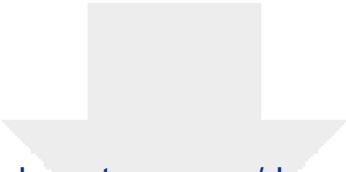
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**Supplementary Interactive Plot Data (CSV)**

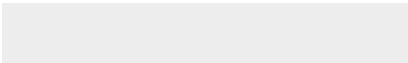
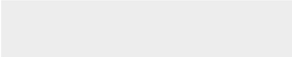
**Supplementary 1.xlsx**







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**Supplementary Interactive Plot Data (CSV)**  
Supplementary Data 2.docx



### **CRedit authorship contribution statement**

**Maarouf Abdou Alio:** Conceptualization, Methodology, Software. Formal analysis, Writing - original draft. **Agnès Pons:** Supervision, Conceptualization, Project administration, Writing - review & editing. **Alain Marcati:** Software, Methodology, Formal analysis. **Christophe Vial:** Software, Methodology, Conceptualization, Supervision, Writing - review & editing, Project administration.

**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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