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## Compared exergy analysis of sugarcane bagasse sequential hydrolysis and fermentation and simultaneous saccharification and fermentation

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**Abstract:** This paper presents the compared exergy analysis of the enzymatic hydrolysis of sugarcane bagasse through Sequential Hydrolysis and Fermentation-SHF and Simultaneous Saccharification and Fermentation-SSF configurations to obtain lignocellulosic ethanol. Four pre-treatment processes were considered: Steam Explosion-SE, Organosolv, Liquid Hot Water-LHW, and a combined SE and LHW method. The pre-treatments analysed simultaneously with the SHF (Cases 1, 3, 5 and 7) and SSF (Cases 2, 4, 6 and 8) configurations were carried out for the overall system focus on the wine production. Processes simulations were performed by using Aspen Plus® to a plant with 500 t/h milling capacity. The results for SHF configurations in terms of exergy efficiency rate were 60.0% for case 1, 56.5% for case 3, 58.3% for case 5 and 59.4% for case 7. For SSF configurations, the exergy efficiency obtained in case 2, case 4, case 6 and case 8 were 62.4%, 58.3%, 60.3% and 61.5%, respectively.

**Keywords:** biomass conversion; bioethanol; exergy efficiency; enzymatic hydrolysis; lignocellulosic biomass; irreversibility rate; sequential hydrolysis and fermentation; SHF; simultaneous saccharification and fermentation; SSF; wine production.

**Reference** to this paper should be made as follows: Silva Ortiz, P. and Oliveira Jr., S. (2016) 'Compared exergy analysis of sugarcane bagasse sequential hydrolysis and fermentation and simultaneous saccharification and fermentation', *Int. J. Exergy*, Vol. 19, No. 4, pp.459–480.

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This paper is a revised and expanded version of a paper entitled 'Compared exergy analysis of sequential enzymatic hydrolysis – fermentation and simultaneous saccharification – fermentation of sugarcane bagasse' presented at *13th International Conference on Clean Energy (ICCE-2014)*, Istanbul, Turkey, 8–12 June, 2014.

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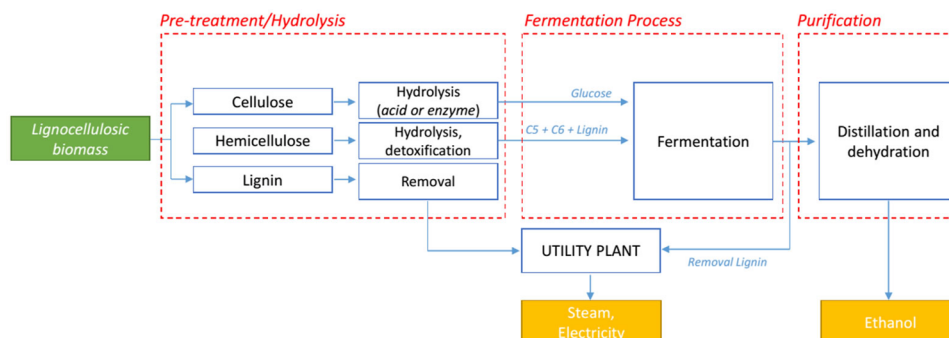
## 1 Introduction

Bioethanol converted from lignocellulosic feedstock, such as sugarcane, is considered one of the technological routes to be developed in the biofuels industry applying the biorefinery concept. Hence, the use of bioethanol as an alternative fuel has become more popular recently due to the oil price volatility and environmental problems associated with fossil fuels (Wirawan et al., 2012). In comparison to fossil-based materials, bio-based feedstock and products have lower carbon efficiency and contrasting molecular structures, and these differences are concerns that must be addressed regarding the biorefineries systems. Therefore, second-generation biofuels produced from lignocellulosic feedstock should be promoted. For instance, the agreement implemented by the Policy Energy Act (PEA) followed by the Energy Independence and Security Act (EISA) aims to reach 36 billion gallons (136.27 billion liters) of bioethanol by the year 2022 (Limayem and Ricke, 2012).

The main stages of the bioethanol production process are pre-treatment, hydrolysis, fermentation, distillation and further fuel upgrading are shown in Figure 1. Lignocellulose must thus be pre-treated before it can be converted to bioethanol. At this stage, the lignocellulose can either be pre-treated using chemical, physical–chemical, physical and biological pre-treatment methods, depending on the desired type of sugar.

After the pre-treatment stage, the lignocellulose can be hydrolysed to produce sugars. There are at least three methods of enzymatic hydrolysis, including dilute acid hydrolysis, concentrated acid hydrolysis and enzymatic hydrolysis. Nowadays, the prevalent technique for bioethanol production is an enzyme-based process because it gives higher hydrolysis yield than acid hydrolysis (Zheng et al., 2009). It is worth noting that the pre-treatment and hydrolysis steps have been highlighted as the most costly stages in the bioprocessing of lignocellulosic materials (Modenbach and Nokes, 2013). For example, pre-treatment as a first step is the most costly operation and accounts for ~33% of the total cost with respect to the economic feasibility of each step (Limayem and Ricke, 2012).

**Figure 1** Processing steps from lignocellulosic biomass feedstocks (see online version for colours)



In this sense, the pre-treatment of the substrates plays an important role because it is costly and affects all downstream operations. The choice and optimisation of pre-treatment techniques has been the focus of many research efforts (Young et al., 2010; Menon and Rao, 2012; Nanda et al., 2014). Hence, the selection of the pre-treatment method largely depends on the type of feedstock, which determines the structure of the lignocellulose biomass. Several pre-treatment approaches have been proposed and tested, but only very few achieve considerable sugar yields with low enough costs (Sánchez and Cardona 2008; Hu and Ragauskas, 2012). The optimisation and integration of pre-treatment methods have remained a challenge and an opportunity to reduce the cost of cellulosic ethanol (Thommes and Strezov, 2014).

In this work, four pre-treatment processes for sugarcane bagasse based on the arrangements analysed by Silva Ortiz and Oliveira (2014) and two bioethanol production configurations (SHF and SSF) were simulated. Thereby, a performance comparison in terms of exergy efficiency and destroyed exergy rate of the cases studied is summarised in Figure 2. Focusing on the outcomes, the impact of SHF and SSF configurations on the average exergy efficiency of the wine production are discussed.

### 1.1 Second-generation bioethanol production

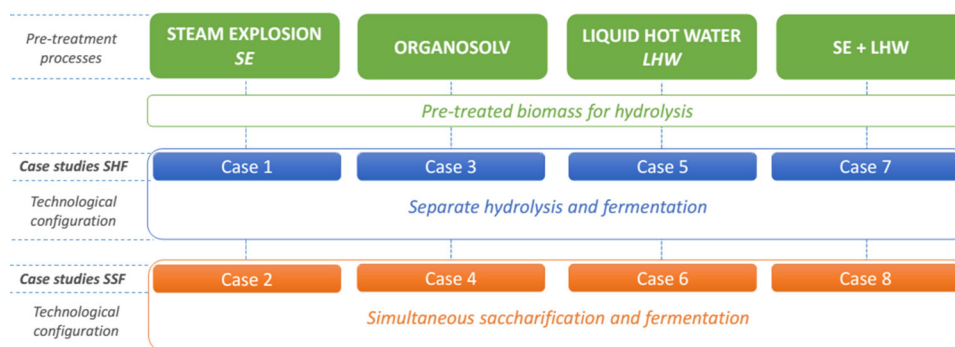
Ethanol production from second-generation (2G) feedstocks still shows significant potential for technical improvements and research efforts have been focused on increasing process efficiencies and reducing costs, as well as water and energy consumption (Process Integration). To reach an economically viable bioethanol production using lignocellulosic materials, each step process has to be optimised so that high ethanol yields can be obtained with the lowest possible cost (Thommes and Strezov, 2014).

Lignocellulosic biomass is considered an attractive feedstock of ethanol production because of its availability in large quantities, at low cost, and its reduced competition with food but not necessarily with feed (Cheng et al., 2008), and taking into account that the raw material constitutes ~40–70% of the production cost (Quintero et al., 2008).

In general, prospective bio-based materials (lignocellulosic feedstock) for bioethanol production can be divided into six main groups: *Crop residues* (sugarcane bagasse, corn stover, wheat straw, rice straw, rice hulls, barley straw, sweet sorghum bagasse, olive stones and pulp); *Hardwood* (aspen, poplar), *Softwood* (pine, spruce); *Cellulose wastes*

(newsprint, waste office paper, recycled paper sludge), *Herbaceous biomass* (alfalfa hay, switch grass, reed canary grass, timothy grass, miscanthus grass) and *municipal solid wastes* (Binod et al., 2011).

**Figure 2** Description of the case studies evaluated (see online version for colours)



### 1.2 Process configurations: saccharification and fermentation route

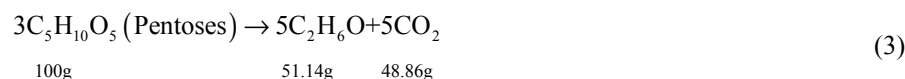
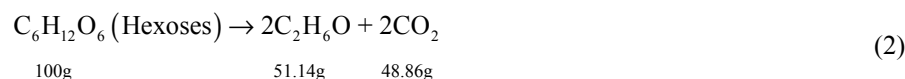
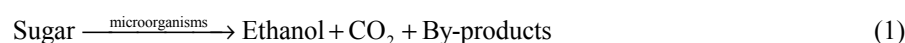
The conventional technologies employed for fermenting biomass hydrolysates involves a sequential process in which the hydrolysis of cellulose and the fermentation are carried out in different units. This configuration is known as separate hydrolysis and fermentation (SHF).

Alternatively, the enzymatic treatment can be accomplished simultaneously with the fermentation process; it is recognised as simultaneous saccharification and fermentation (SSF), in which hydrolysis and fermentation are performed in a single unit. The most employed microorganism in corn-based and sugar-based biofuel industries for fermenting lignocellulosic hydrolysates is *Saccharomyces cerevisiae* (*S. cerevisiae*). The yeast *S. cerevisiae* is used as the ethanol-producing microorganism in almost all industrial-scale plants in Brazil. An overview of main operational conditions for this yeast (microorganism) can be found in Table 1.

**Table 1** Saccharification-fermentation reactions and conversion rates

Yeast	<i>T</i> (°C)	<i>pH</i> range	Carbon source (Sugar)	Ethanol yield (%)	Application and status
<i>S. cerevisiae</i>	28–32	3–5	Glucose, fructose, sucrose, galactose, mannose, maltose	73–94	Industrial scale: sugarcane, corn, wheat

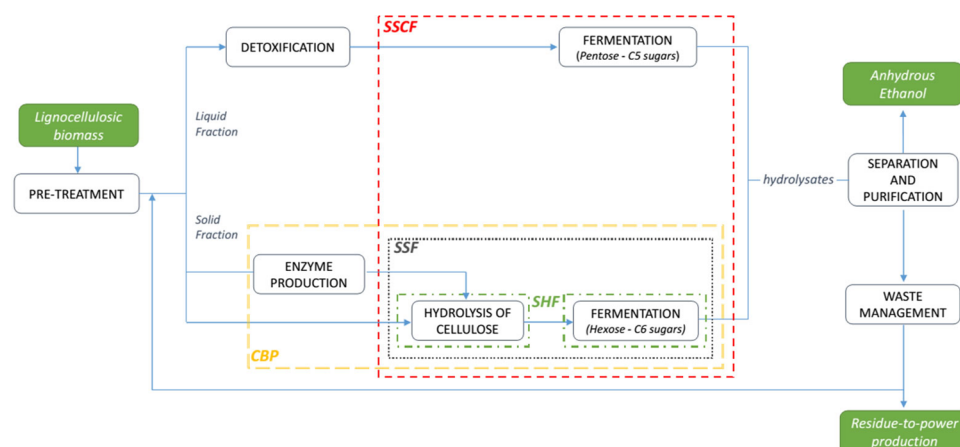
This yeast ferments the hexoses contained in the hydrolysate but not the pentoses. *S. cerevisiae* can generate a high yield of ethanol (12.0–17.0% w/v; 90% of the theoretical value) from hexose sugars (Limayem and Ricke, 2012). The general overall reaction of the fermentation process is presented from equations (1) to (3). The reactions involved in pentose (five-carbon chain, C5) sugar conversion are significantly slower than those for hexoses (six-carbon chain, C6) and only a few microorganisms have been found to be able to produce ethanol from pentose sugars. Therefore, industrial applications have thus far been limited (Thommes and Strezov, 2014).



It must be pointed out that ethanol is produced in large scale from sugarcane in Brazil by fermentation of sugars and distillation (first-generation, 1G). Currently, this process is considered an efficient biofuel technology, leading to significant reduction in greenhouse gases emissions. However, some improvements in the process can be introduced in order to reduce the use of energy. In current distilleries, a significant fraction of the energy consumption occurs at the purification step (distillation and dehydration) since conventional fermentation systems employed in the industry require low substrate concentration, which must be distilled, consequently with high energy consumption. Alternatives to the conventional fermentation processes were assessed in this regard, through computer simulation for low temperature fermentation and vacuum extractive fermentation arrangements. Results show that the ethanol production increases between 3.3% and 4.8% and a reduction of up to 36% in steam consumption occurs (Palacios-Bereche et al., 2014).

Another technological alternative is the integration of the pentose fermentation in the process. This route is known as simultaneous saccharification and co-fermentation (SSCF). A scheme of bioethanol production processes from lignocellulosic biomass is given in Figure 3. Finally, consolidated bioprocessing (CBP), features the combination of cellulase production, enzymatic cellulose hydrolysis and fermentation in a single process step.

**Figure 3** Configurations of processes of bioethanol production from sugarcane bagasse (see online version for colours)



Source: Adapted from Cardona et al. (2010)

### 1.2.1 *Separate (or sequential) hydrolysis and fermentation (SHF)*

In this traditional method, the hydrolysis and fermentation are two distinct process steps and the fermentation of hexoses and pentoses is carried out in separate reactors. When the SHF configuration is used, the solid fraction of pre-treated lignocellulosic material undergoes hydrolysis (saccharification).

This fraction contains the cellulose in an accessible form to acids or enzymes. Once hydrolysis is completed, the resulting cellulose hydrolysate is fermented and converted into ethanol. One of the main features of the SHF process is that each step can be performed at its optimal operating conditions. The most important factors to consider for the saccharification step are reaction time, temperature, pH, enzyme dosage and substrate load.

Tests on lignocellulosic biomass from sugarcane leaves have found the best values of all these parameters and 65–70% cellulose conversion was achieved at 50°C and a pH of 4.5. Although enzyme doses of 100 FPU/g cellulose resulted in close to 100% hydrolysis, this amount of cellulases enzymes is not economically justifiable. Hence, 40 FPU/g cellulose dosage was proposed obtaining only 13% reduction in conversion (Sánchez and Cardona, 2008). Regarding the substrate concentration, solids loads of 10% were defined as the most adequate considering mixing difficulties and accumulation of inhibitors in the reactive medium.

Lastly, some examples of bioethanol plants that utilise SHF process are the Canadian company, *Iogen Corporation*, and the Spanish company, *Abengoa*, both using straw as the biomass feedstock (Thommes and Strezov, 2014).

### 1.2.2 *Simultaneous saccharification and fermentation (SSF)*

The SSF process reducing sugars produced in cellulose hydrolysis are simultaneously fermented to ethanol, which greatly reduces the product inhibition of hydrolysis. This process shows more attractive indexes than SHF, such as higher ethanol yields and less energy consumption. In this case, cellulase enzymes and microorganisms are added to the same reactor allowing the glucose formed during the enzymatic hydrolysis of cellulose to be immediately consumed by the microbial cells converting it into ethanol.

However, the need of employing more diluted media to reach suitable rheological properties makes the final product concentration be low. In addition, this process operates at non-optimal conditions for hydrolysis and requires higher enzyme dosage, which influences substrate conversion positively but process costs negatively. Hence, considering that enzymes account for an important share of production costs, it is necessary to find methods to reduce the cellulases doses to be used (Sánchez and Cardona, 2008).

In this process configuration, the optimal temperature for yeasts is usually around 30°C, but it can be increased by using thermo-tolerant yeasts. The hydrolysis optimum is found between 45°C and 50°C and most often a compromise of ~ 38°C is used in the SSF process (Thommes and Strezov, 2014).

### 1.2.3 *Simultaneous saccharification and co-fermentation (SSCF)*

Another technological alternative is the integration of the pentose fermentation (liquid stream containing C5 sugars) in the SSF route as shown in Figure 3. This configuration is accomplished by combining the enzymatic hydrolysis of cellulose to the glucose process

and the co-fermentation of pentose and hexose sugars process in one reaction vessel. Both fermenting microorganisms have to be compatible in terms of operating pH and temperature. This reduces the number of reactors involved by eliminating the separate hydrolysis reactor and avoids the problem of product inhibition associated with enzymes (Hamelinck et al., 2005).

Some drawbacks of the SSCF configuration are the high by-product formation in the form of carbon dioxide-CO<sub>2</sub> and xylitol-C<sub>5</sub>H<sub>12</sub>O<sub>5</sub>, poor enzyme stability, incompatible pH and temperature (pH of 7.0 and 70°C for the isomerisation process), and the reversibility of the enzyme transformation (Sánchez and Cardona, 2008). A process challenge in the biotechnological conversion of lignocellulosic material in these technological pathways is the efficient conversion of all sugars (C5 and C6) into bioethanol, especially for hardwoods feedstock which have greater amounts of pentoses (Binod et al., 2011).

#### 1.2.4 Consolidated bioprocessing (CBP)

In consolidated bioprocessing, the hydrolysis and fermentation steps and the enzyme production are carried out in a single reactor vessel. One microorganism or a combination of microorganisms able to produce the required enzymes for hydrolysis and convert the fermentable sugars into ethanol is used. No organism having all the desirable features for this processing scheme has been found in nature (Thommes and Strezov, 2014).

The concept of CBP involves the following four biologically mediated transformations:

- the production of saccharolytic enzymes (cellulases and hemicellulases)
- the hydrolysis of carbohydrate components present in pre-treated biomass to form sugars
- the fermentation of hexose sugars (glucose, mannose and galactose)
- the fermentation of pentose sugars (xylose and arabinose) (Singhania et al., 2013).

CBP has been recognised as a potential method for economic bioethanol production from biomass, and intensive research has been undertaken in this area (Thommes and Strezov, 2014).

A model developed at the National Renewable Energy Laboratory (NREL) to analyse advances in biomass processing systems indicated that increasing levels of process consolidation can result in greater cost reductions, and CBP offers the largest cost reduction of any process improvement considered to date (Aden et al., 2013). However, this technique faces the problem of the low tolerance of clostridia (highly polyphyletic class of Firmicutes) to ethanol and the reduction in the ethanol yield due to the formation of acetic acid and salts of other organic acids like lactates (Singhania et al., 2013).

## 2 Processes description

Four pre-treatment processes for sugarcane bagasse based on the technologies analysed by Silva Ortiz and Oliveira (2014) and two bioethanol production configurations (SHF and SSF) were simulated, using Aspen Plus<sup>®</sup> software to a plant with 500 t/h milling capacity considering steady-state conditions for all cases. Process simulations to

evaluate mass, energy and exergy balances were performed. Regarding convergence in simulation, the Wegstein method was used by the simulator (default method in Aspen Plus®) to carry out mass and energy balances in each operation.

Eight case studies combined SHF and SSF configurations with the following pre-treatment technologies are studied: Steam Explosion (SE), Organosolv, Liquid Hot Water (LHW) and SE + LHW. These processes are the most widely used as pre-treatment methods when sugarcane bagasse is the feedstock (Hamelinck et al., 2005; Carrasco et al., 2010; Dias et al., 2012).

It is worth highlighting that enzymatic hydrolysis is usually catalysed by cellulase enzymes and the fermentation is carried out by yeast or bacteria. Cellobiose and glucose are known to inhibit the activities of cellulase enzymes even at low concentrations. When SHF is performed, enzymatic hydrolysis and fermentation are run in two different vessels. In this process, each step can be conducted at optimal conditions of pH and temperature. However, glucose and cellobiose accumulation in the hydrolysis step inhibits the activity of the cellulase enzymes.

In a SSF process, the enzymatic hydrolysis and fermentation are run in the same vessel. Thus, glucose released by the action of cellulase enzymes is converted directly into ethanol by the fermenting microorganism and this continuous removal of glucose from the medium minimises the end-product inhibition on enzyme activity (Ojeda et al., 2011a).

Moreover, case studies represent steps of the integrated process for 1G and 2G bioethanol and electricity production. In this configuration, sugarcane bagasse and trash are used as fuels in the cogeneration system (utility plant), and the amount that exceeds the one required to supply the thermal energy demand of the plant is used as feedstock in the second-generation ethanol production process. A simplified diagram of the integrated process is shown in Figure 4, where the dashed lines in red express the control volume considered in this analysis. Consequently, the focus of the studies was wine production.

As shown in this diagram, residues of the 2G process are also used as fuels in the utility plant. The unreacted solids (cellulose and lignin) are fed to the boilers. In this particular case, pre-treatment technologies are employed in order to remove bagasse lignin prior to enzymatic hydrolysis, and the unreacted solids obtained after removal of the glucose liquor produced in the enzymatic hydrolysis reactor contain mainly cellulose. Besides, pentoses released during pre-treatment can be either biodigested, producing biogas which is also burnt to produce energy, or fermented into ethanol.

## 2.1 Raw material

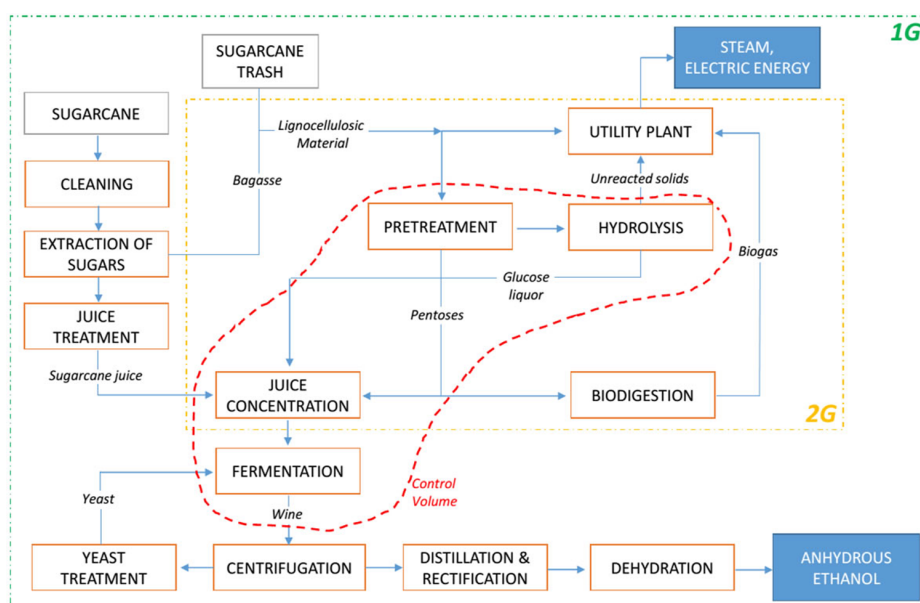
For all cases, sugarcane bagasse is the feedstock considered in the simulations performed. This lignocellulosic material is assumed to have the following typical chemical normalised composition (w/w): *Cellulose* 43.38%, *Hemicellulose* 25.63%, *Lignin* 23.24%, *Ash* 2.94% and *Extractives* 4.81%. The extractives were represented as organic acids. These components were inserted into the simulator with molecular formula  $C_6H_6O_6$ . The moisture content of the bagasse produced in the mills was set as 50%.

This average composition is reported in Rocha et al. (2010), in which 50 samples of bagasse from mills located in different parts of Brazil and in different seasons



were measured. The properties of the components of the lignocellulosic material were obtained from the databank for biofuel components developed by NREL (Wooley and Putsche, 1996).

**Figure 4** Scheme for integrated 1G-2G ethanol and electricity production (see online version for colours)



Source: Adapted from Dias et al. (2013).

## 2.2 Reactor models

Stoichiometric reactors (RStoic model) were used in the simulator to represent the reactions involved in the hydrolysis–saccharification (R-HYDRO) and fermentation processes (R-FERM). Table 2 shows the reactions and conversion yields considering 10% of solids content in the hydrolysis reactor, according to NREL (Humbird et al., 2011). Furthermore, this table indicates the formation of the main components in the reactors.

Table 3 provides the operating conditions defined for the SHF and SSF processes. For each configuration, water was added in order to remove xylose and inhibitor components, as well as to adjust the concentration of water-insoluble solids in the hydrolysis reactor. The water evaluated for the washing step (H2O-WASH) was 123.4 kg/s, at a temperature of 25°C and a pressure of 101.325 kPa; these were adopted as input parameters in the simulations. Meanwhile, the operating conditions defined for water of the enzymatic hydrolysis of bagasse (H2O-HYD) was 103 kg/s, at a temperature of 25°C and a pressure of 250 kPa, based on the operational parameters of the integrated 1G and 2G ethanol production from sugarcane reported by Palacios-Bereche et al. (2013).

Additionally, cellulase enzyme in the reactors as bioagent was adopted, according to the processes analysed, as per Ojeda et al. (2011a). For the simulations, an average value of 0.114 g enzyme/g dry pre-treated pulp was considered as in Carrasco et al. (2010).

**Table 2** Saccharification-fermentation reactions and conversion rates

<i>Reactions</i>	<i>Yield conversion (%)</i>
<i>Saccharification</i>	
CELLULOSE (C <sub>6</sub> H <sub>10</sub> O <sub>5</sub> ) + WATER (H <sub>2</sub> O) → GLUCOSE (C <sub>6</sub> H <sub>12</sub> O <sub>6</sub> )	55.8
HEMICELLULOSE (C <sub>5</sub> H <sub>8</sub> O <sub>4</sub> ) + WATER (H <sub>2</sub> O) → XYLOSE (C <sub>5</sub> H <sub>10</sub> O <sub>5</sub> )	40.6
<i>Fermentation</i>	
GLUCOSE (C <sub>6</sub> H <sub>12</sub> O <sub>6</sub> ) → 2 ETHANOL (C <sub>2</sub> H <sub>6</sub> O) + 2 CO <sub>2</sub>	90.0
GLUCOSE (C <sub>6</sub> H <sub>12</sub> O <sub>6</sub> ) + 2 WATER (H <sub>2</sub> O) → 2 GLYCEROL (C <sub>3</sub> H <sub>8</sub> O <sub>3</sub> ) + O <sub>2</sub>	0.4
GLUCOSE (C <sub>6</sub> H <sub>12</sub> O <sub>6</sub> ) + 2 CO <sub>2</sub> → 2 SUCCINIC ACID (C <sub>4</sub> H <sub>6</sub> O <sub>4</sub> ) + O <sub>2</sub>	0.6
3 XYLOSE (C <sub>5</sub> H <sub>10</sub> O <sub>5</sub> ) → 5 ETHANOL (C <sub>2</sub> H <sub>6</sub> O) + 5 CO <sub>2</sub>	80.0
3 XYLOSE + 5 CO <sub>2</sub> → 5 ACETIC ACID (C <sub>2</sub> H <sub>4</sub> O <sub>2</sub> ) + 2.5 O <sub>2</sub>	0.9

**Table 3** Saccharification-fermentation operational conditions

<i>Parameter</i>	<i>SHF</i>	<i>SSF</i>
Cellulase enzyme	15 FPU/g	15 FPU/g
Pressure	101.325 kPa	101.325 kPa
Temperature	321 K	308 K
Bioagent	<i>S. cerevisiae</i>	<i>S. cerevisiae</i>

In each case analysed, the pre-treated biomass was sent to SHF (Figure 6) and SSF (Figure 7) configuration, respectively. At first, the mixture composed of the pre-treated bagasse and the washing water, B-PRET stream, goes through a separated unit (SEPA-PE) to split the liquid fraction (PEN-LIQ) xylose liquor (pentose) from the solid fraction of cellulose and lignin (CEL-LIG). In this study, the use of xylose liquor was not considered.

For the next stage, water is added to the process (stream H<sub>2</sub>O-HYD) in tank T-MIST, to achieve an appropriate concentration of water insoluble solids in the hydrolysis reactor. Afterwards, stream CEL-LIG2 goes to the hydrolysis reactor (R-HYDRO), where enzymes are added to catalyse the simultaneous reactions involved in this process. After the hydrolysis stage in both models, the hydrolysate goes through a filter in order to separate the lignin cake (solid fraction) from the glucose liquor (GLU-LIQ).

Before the glucose liquor can proceed to fermentation, it needs to be concentrated. This process takes place in a multiple-effect evaporation system. The hierarchy model (CONC) was performed in Aspen Plus<sup>®</sup> to represent this process. Figure 5 provides an overview of the parameters evaluated in the multiple-effect evaporation system.

Initially, the hydrolysate (GLU-LIQ) is preheated before entering the evaporation system, which operates with exhaust steam at 2.5 bar. A five-stage evaporation system was assumed to reduce steam consumption. Each stage of the evaporation system was considered to have two components, a heat exchanger and a flash separator. The evaluated steam flow (STEAM-EV) was 25.9 kg/s, a temperature of 127°C and a pressure of 2.5 bar based on Palacios-Bereche et al. (2013). The products obtained from the CONC model were juice vapour (JUICEVAP) condensates of exhaust steam

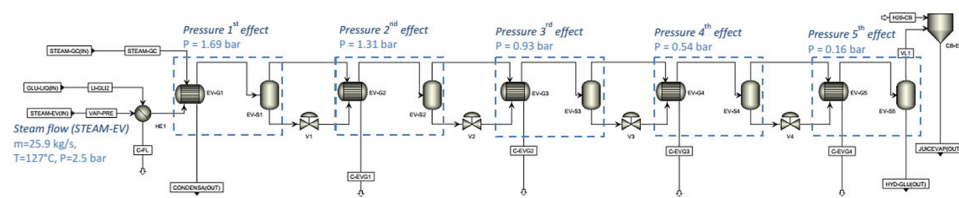
(CONDESA) and glucose hydrolysate (HYD-GLU). Lastly, the solid content in concentrate hydrolysate was 17%.

It must be pointed out that due to the possible presence of soluble lignin in glucose hydrolysate, as well as phenolic groups, the HYD-GLU steam is prepared via detoxification before performing the fermentation process. In this study, fermentation was based on the Melle-Boinot process (cell-recycle batch) to ferment the xylose. In a conventional autonomous distillery, vapour bleedings and condensates with different pressures and temperatures resulting from the concentration of the glucose process are used in the evaporation system and as input for other processes.

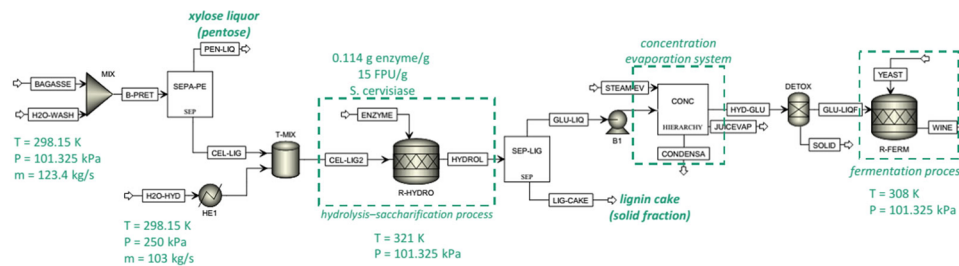
Figure 6 shows the schematic representation of the SHF process. This configuration is divided into two reactors. The first one represents the hydrolysis reactor (R-HYDRO) and the second one the sugars fermentation reactor (R-FERM).

A scheme and the characteristics of SSF configuration on Aspen Plus<sup>®</sup> software is given in Figure 7, where saccharification (hydrolysis) and fermentation processes occur simultaneously in the reactor (R-HYDFER).

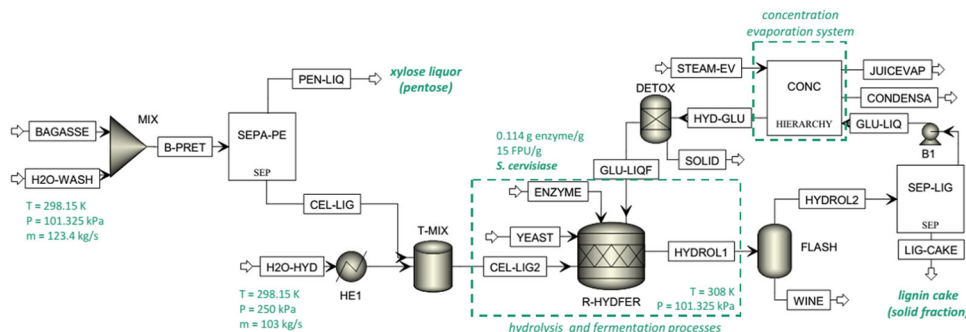
**Figure 5** Scheme for the evaporation system-specifications of the concentration process (see online version for colours)



**Figure 6** Flow sheet of the SHF configuration (see online version for colours)



**Figure 7** Flow sheet of the SSF configuration (see online version for colours)

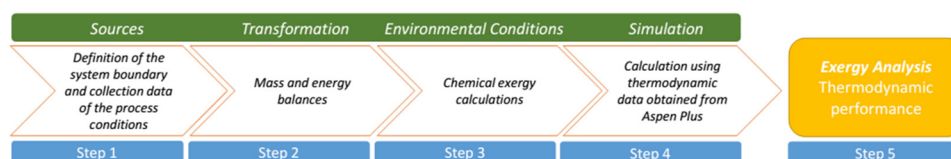


### 3 Exergy analysis

This method consists in using the First and Second laws of Thermodynamics together for furthering the goal of more effective energy resource use, for it allows the location, cause, and true magnitude of waste and loss to be determined. Such information can be used to design new energy efficient systems and to increase the efficiency of existing systems (Bejan et al., 1996).

The exergy analysis was carried out to develop the procedure based on the methodology proposed in Silva Ortiz and Oliveira (2014). In this sense, a Microsoft Excel<sup>®</sup> tool was developed to perform the exergy analysis for the proposed routes. This application is necessary because Aspen Plus<sup>®</sup> does not have an integrated function to calculate the exergy of each stream presented in the processes, or to evaluate thermo-economic costs, although it gives enough thermodynamic data and can also estimate capital costs. In order to calculate the exergy of matter streams, some authors have approached it in several ways, including Fortran subroutines (Mosqueira-Salazar et al., 2013; Palacios-Bereche et al., 2013), compiled applications within the Aspen Plus<sup>®</sup> interface (Ojeda et al., 2011a, 2011b), a Microsoft Excel<sup>®</sup> VBA supplement (Querol et al., 2011) or hand-made calculations. The systematic procedure involved in the assessment of SHF and SSF routes via exergy analysis is shown in Figure 8.

**Figure 8** Procedure for exergy analysis of SHF and SSF processes (see online version for colours)



Different process configurations have been investigated to apply and to develop an efficient conversion process for second-generation ethanol production (Dias et al., 2012, 2013; Quintero and Cardona, 2011; Sánchez and Cardona, 2012; Panwar, 2014).

For the routes proposed, thermodynamic data of the streams and substances present in the processes are necessary. The exergy analysis throughout this work was conducted in Aspen Plus<sup>®</sup>, using the data from the different matter streams: mass flow rate, temperature, pressure and composition. The enthalpy and entropy of the streams with the same composition are also obtained, considering that they are at ambient temperature and pressure. Thus, a conventional reference environment is established, at  $T_0 = 25^\circ\text{C}$  and  $p_0 = 101.325\text{ kPa}$ , since most of the thermodynamic properties are tabulated at  $T_0, p_0$ .

#### 3.1 Equations used for energy and exergy performance

Note that the total specific exergy is calculated as in Szargut et al. (1988), as the sum of physical and chemical components. Thus, the physical exergy was determined according to equation (4) in order to calculate enthalpy and entropy in actual and reference conditions.

$$b_{ph} = (h - h_0) - T_0 \cdot (s - s_0) \quad (4)$$

Furthermore, the chemical exergy calculation for each component was estimated using Szargut et al. (1988), as indicated in Silva Ortiz and Oliveira (2014).

The standard chemical exergy of pure components were obtained from Szargut et al. (1988) and Ojeda et al. (2011a). However, data were not available for some components such as aconitic acid ( $C_6H_6O_6$ ), glycerol ( $C_3H_8O_3$ ), furfural ( $C_5H_4O_2$ ), xylose ( $C_5H_{10}O_5$ ), sulphurous acid ( $H_2SO_3$ ), yeast ( $CH_{1.8}O_{0.9}N_{0.1}$ ) and enzymes ( $CH_{1.57}N_{0.29}O_{0.31}S_{0.007}$ ). Aconitic acid is found in sugarcane, and glycerol is a by-product of fermentation. Furfural and xylose appear in the pre-treatment of sugarcane bagasse from the hydrolysis of hemicelluloses.

For sulphurous acid, standard chemical exergy was evaluated based on Palacios-Bereche et al. (2013). For aconitic acid, furfural, glycerol, xylose, yeast and enzymes, the standard chemical exergy was calculated according to the technical fuels procedure based on net calorific values and atomic ratios (Szargut et al., 1988). For the cellulase enzyme, the composition was assumed, as per Wooley and Putsche (1996). The chemical exergies of the substances involved in the processes are listed in Table 4.

**Table 4** Specific chemical exergy of the substances studied in the processes

Formula	Substance	Component ID Aspen Plus <sup>®</sup>	$b_{CH}^{specific}$ (kJ/kg)
$C_6H_{10}O_5$	CELLULOSE	CELU	18,875
$C_5H_8O_4$	HEMICELLULOSE	HEMI	19,177
$C_{7.3}H_{13.9}O_{1.3}$	LIGNIN	LIGN	37,133
$C_{12}H_{22}O_{11}$	SUCROSE	C12H22O11	18,185
$C_6H_{12}O_6$	GLUCOSE	C6H12-01	17,583
$K_2O$	POTASSIUM OXIDE	K2O	4395
KCL	POTASSIUM CHLORIDE	KCL	261
$H_2O$	WATER (L)	H2O	50
$SiO_2$	SILICON DIOXIDE	SIO2	59*
$C_5H_{10}O_5$	XYLOSE	C5H10-01	17,583
$C_5H_4O_2$	FURFURAL	C5H4O-01	24,437
$H_2SO_3$	SULFUROUS ACID	H2SO3	3,962
$CH_{1.57}N_{0.29}O_{0.31}S_{0.007}$	ENZYME	ENZYME	24,967
$CH_{1.8}O_{0.9}N_{0.1}$	YEAST	YEAST	17,350
$C_2H_6O$	ETHANOL	C2H6O-2	29,471
$CO_2$	CARBON DIOXIDE	CO2	451
$C_4H_6O_4$	SUCCINIC ACID	C4H6O-01	15,949
$C_2H_4O_2$	ACETIC ACID	C2H4O-01	17,583
$C_5H_{12}O$	AMYL ALCOHOL	C5H12O-03	38,389
$C_3H_8O_3$	GLYCEROL	C3H8O-01	19,667
$NH_3$	AMMONIA	NH3	19,876
$C_6H_6O_6$	ORGANIC ACIDS	C6H6O-01	14,275

\*To determine the specific chemical exergy of  $SiO_2$ , a content of amorphous material of 25% was considered. Besides, the proportions of cristobalite and quartz were assumed to have 16% and 59% (in mass), according to the parameters for sugarcane bagasse ash reported in Cordeiro et al. (2008).

Finally, the quality of the energy conversion processes is quantified by the exergy efficiency given by equation (5), and the irreversibility rate (equation (6)).

$$\eta_B = \frac{\sum B_{\text{products}}}{\sum B_{\text{inputs}}} \quad (5)$$

$$I = \sum B_{\text{inputs}} - \sum B_{\text{products}} \quad (6)$$

#### 4 Results and discussion

Exergy analysis is fundamental for accounting both for materials use and waste residuals. In the exergy analysis performed in this paper, the entropy generation rate (destroyed exergy rate) and the exergy efficiency were calculated for each case study establishing a relationship between them seeking to improve the configurations efficiency and sustainability.

Tables 5–8 show the values of some parameters used in the evaluation of the SHF and SSF technologies. The results show that all cases of separate saccharification and fermentation (SHF) configuration have the lowest exergy efficiencies. One of the main reasons for these irreversibilities is the inhibition effect of glucose and cellobiose accumulation in the SHF reactor. Hence, the inhibitor substantially reduces the enzyme velocity at low substrate concentrations reducing the exergy efficiency of the processes.

The present findings also suggest that some improvements in wine production processes via enzymatic technology, SHF and SSF configurations, using pre-treated bagasse. It is evident that wine production for the cases studied are a function of the mass flow inlet of pre-treated sugarcane bagasse as illustrated in Table 6 for cases 3 and 4.

A sensitivity analysis for the cases studied was performed, varying the mass flow rate of the enzyme in order to evaluate their impact on wine production. The results show that it does not represent a significant fraction (<2%) for increasing the total wine production for all cases. For this reason, mass flow rate of the enzyme was fixed in the configurations. In fact, the cost of enzymes is one of the most important challenges in cellulosic ethanol production. Thus, technological pathways proposed in this matter have addressed concerns to achieve the rational use of resources, especially with regard to the identification of optimal dosages for the enzymes.

Furthermore, cases 1 and 2 represent the best option regarding the production of xylose liquor (pentose). This product can be used as a main input for an up flow anaerobic sludge blanket reactor to produce biogas from the organic matter present in it.

The higher exergy destruction and losses in enzymatic hydrolysis found in cases 3 and 4 are due to the high enzyme consumption and the amount of bagasse to be hydrolysed (Figure 10). Moreover, the efficiency of the hydrolysis configurations is associated to the pre-treatment processes of bagasse and the reactions occurring in the reactors, where sugar recovery by enzymatic attack is not complete.

**Table 5** Inlet and outlet parameters of mass flow rates of cases 1 and 2 (SE)

<i>Parameter Streams</i>		<i>Configuration</i>	<i>Mass flow</i> (kg/s)	<i>T</i> (K)	<i>P</i> (kPa)	<i>b</i> (kJ/kg)	<i>B<sub>tot</sub></i> (kW)
Inlets	PRETREATED BAGASSE	SHF, SSF	16.94	298.15	101,325	10,570	179,056
	ENZYME	SHF, SSF	0.60	302.15	101,325	23,730	14,238
	H2O-HYD	SHF, SSF	135.15	323.15	250	54.3	7339
	H2O-WASH	SHF, SSF	161.93	298.15	101,325	50	8096
	STEAM-EV	SHF, SSF	33.98	400.15	250	110.3	3748
	YEAST	SHF, SSF	3.54	304.15	101,325	1177	4167
	<i>TOTAL</i>	<i>SHF, SSF</i>	<i>352.14</i>				<i>216,640</i>
Outlets	LIG-CAKE	SHF	11.14	323.15	101,325	1226	13,658
		SSF	15.14			1072	16,230
	PEN-LIQ	SHF	120.58	310.15	101,325	138	16,640
		SSF	121.46			140	17,004
	SOLID	SHF	3.91	323.15	101,325	1298	5075
		SSF	5.71			1135	6481
	WINE	SHF	124.39	304.15	101,325	706	87,819
		SSF	126.15			708	89,314
	CONDENSA	SHF	30.90	400.15	250	110.3	3408
		SSF	29.90			110.3	3298
	JUICEVAP	SHF	61.22	329.15	50	56.3	3447
		SSF	53.78			56.3	3028
	<i>TOTAL</i>	<i>SHF</i>	<i>352.14</i>				<i>130,050</i>
		<i>SSF</i>	<i>352.14</i>				<i>135,360</i>

**Table 6** Inlet and outlet parameters of mass flow rates of cases 3 and 4 (Organosolv)

<i>Parameter Streams</i>		<i>Configuration</i>	<i>Mass flow</i> (kg/s)	<i>T</i> (K)	<i>P</i> (kPa)	<i>b</i> (kJ/kg)	<i>B<sub>tot</sub></i> (kW)
Inlets	PRETREATED BAGASSE	SHF, SSF	21.97	298.15	101.325	10,570	232,223
	ENZYME	SHF, SSF	0.60	302.15	101.325	23,730	14,238
	H2O-HYD	SHF, SSF	175.28	323.15	250	54.3	9518
	H2O-WASH	SHF, SSF	210	298.15	101.325	50	10,500
	STEAM-EV	SHF, SSF	44.09	400.15	250	110.3	4862
	YEAST	SHF, SSF	3.54	304.15	101.325	1,177	4167
	<i>TOTAL</i>	<i>SHF, SSF</i>	<i>455.48</i>				<i>275,510</i>

**Table 6** Inlet and outlet parameters of mass flow rates of cases 3 and 4 (Organosolv)  
(continued)

<i>Parameter Streams</i>		<i>Configuration</i>	<i>Mass flow (kg/s)</i>	<i>T (K)</i>	<i>P (kPa)</i>	<i>b (kJ/kg)</i>	<i>B<sub>tot</sub> (kW)</i>	
Outlets	LIG-CAKE	SHF	14.39	323.15	101.325	1015	14,606	
		SSF	13.39			1115	14,930	
	PEN-LIQ	SHF	115.41	310.15	101.325	218	25,159	
		SSF	117.66			238	28,003	
	SOLID	SHF	5.80	323.15	101.325	1030	5974	
		SSF	4.80			1130	5424	
	WINE	SHF	127.39	304.15	101.325	741	94,396	
		SSF	129.21			749	96,778	
	CONDENSA	SHF	89.23	400.15	250	110.3	9842	
		SSF	89.23			110.3	9842	
	JUICEVAP	SHF	103.26	329.15	50	56.3	5814	
		SSF	101.19			56.3	5697	
	<i>TOTAL</i>		<i>SHF</i>	<i>455.48</i>				<i>155,790</i>
			<i>SSF</i>	<i>455.48</i>				<i>160,670</i>

**Table 7** Inlet and outlet parameters of mass flow rates of cases 5 and 6 (LHW)

<i>Parameter Streams</i>		<i>Configuration</i>	<i>Mass flow</i> (kg/s)	<i>T</i> (K)	<i>P</i> (kPa)	<i>b</i> (kJ/kg)	<i>B<sub>tot</sub></i> (kW)
Inlets	PRETREATED BAGASSE	SHF, SSF	12.91	298.15	101.325	10,570	136,459
	ENZYME	SHF, SSF	0.60	302.15	101.325	23,730	14,238
	H2O-HYD	SHF, SSF	103	323.15	250	54.3	5593
	H2O-WASH	SHF, SSF	123.40	298.15	101.325	50	6170
	STEAM-EV	SHF, SSF	25.90	400.15	250	110	2849
	YEAST	SHF, SSF	3.54	304.15	101.325	1177	4167
	<i>TOTAL</i>	<i>SHF, SSF</i>	<i>269.36</i>				<i>136,490</i>
Outlets	LIG-CAKE	SHF	8.77	323.15	101.325	1145	10,042
		SSF	9.39			1006	9446
	PEN-LIQ	SHF	102.75	310.15	101.325	101	10,378
		SSF	105.34			102	10,745
	SOLID	SHF	1.28	323.15	101.325	1178	1508
		SSF	3.07			1024	3144
	WINE	SHF	113.46	304.15	101.325	645	73,182
		SSF	115.18			661	76,141



**Table 7** Inlet and outlet parameters of mass flow rates of cases 5 and 6 (LHW) (continued)

<i>Parameter Streams</i>		<i>Configuration</i>	<i>Mass flow</i> (kg/s)	<i>T</i> (K)	<i>P</i> (kPa)	<i>b</i> (kJ/kg)	<i>B<sub>tot</sub></i> (kW)
Outlets	CONDENSA	SHF	24.90	400.15	250	110.3	2746
		SSF	25.90			110.3	2857
	JUICEVAP	SHF	18.20	329.15	50	56.3	1025
		SSF	10.48			56.3	5900
<i>TOTAL</i>		<i>SHF</i>	<i>269.36</i>				<i>98,810</i>
		<i>SSF</i>	<i>269.36</i>				<i>108,233</i>

**Table 8** Inlet and outlet parameters of mass flow rates of cases 7 and 8 (SE+LHW)

<i>Parameter Streams</i>		<i>Configuration</i>	<i>Mass flow</i> (kg/s)	<i>T</i> (K)	<i>P</i> (kPa)	<i>b</i> (kJ/kg)	<i>B<sub>tot</sub></i> (kW)	
Inlets	PRETREATED BAGASSE	SHF, SSF	12.96	298.15	101.325	10,570	136,987	
	ENZYME	SHF, SSF	0.60	302.15	101.325	23,730	14,238	
	H2O-HYD	SHF, SSF	103.40	323.15	250	54.3	5615	
	H2O-WASH	SHF, SSF	123.88	298.15	101.325	50	6194	
	STEAM-EV	SHF, SSF	26	400.15	250	110	2860	
	YEAST	SHF, SSF	3.54	304.15	101.325	1,177	4167	
<i>TOTAL</i>		<i>SHF, SSF</i>	<i>270.38</i>				<i>170,060</i>	
Outlets	LIG-CAKE	SHF	8.42	323.15	101.325	1,008	8487	
		SSF	8.42			1,054	8875	
	PEN-LIQ	SHF	110.70	310.15	101.325	116	12,841	
		SSF	113.70			128	14,554	
	SOLID	SHF	1.91	323.15	101.325	1,022	1952	
		SSF	1.91			1,062	2028	
	WINE	SHF	116.73	304.15	101.325	645	75,291	
		SSF	118.73			649	77,056	
	CONDENSA	SHF	22.12	400.15	250	110.3	2440	
		SSF	19.12			110.3	2109	
	JUICEVAP	SHF	10.50	329.15	50	56.3	5910	
		SSF	8.50			56.3	4790	
	<i>TOTAL</i>		<i>SHF</i>	<i>270.38</i>				<i>106,920</i>
			<i>SSF</i>	<i>270.38</i>				<i>109,410</i>

These results can be compared with others in the literature. Ojeda et al. performed an application of computer-aided process engineering and exergy analysis to evaluate different routes of bioethanol production from lignocellulosic biomass applying the SSF configuration. The reported exergy efficiency for the case study using steam explosion

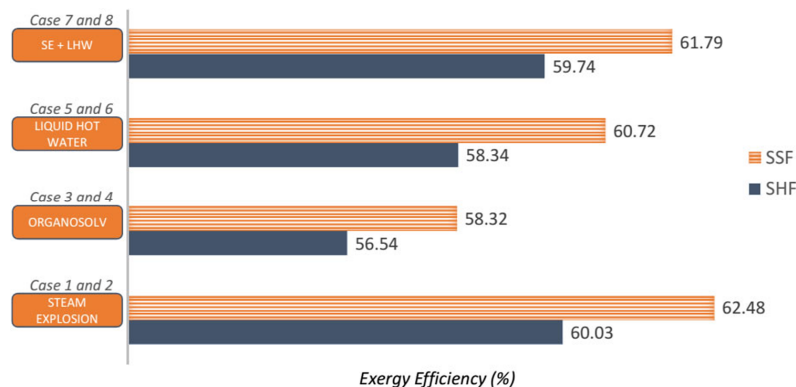
pre-treatment (case 2) was 62.85% and for Organosolv method (case 4) it was 58.09% (Ojeda et al., 2011c).

Ojeda et al. studied ethanol production by enzymatic hydrolysis of sugarcane bagasse. These authors adopted an acid-catalysed steam explosion pre-treatment, pentose fermentation, SSF for cellulose, distillation and rectification, and molecular sieves for anhydrous ethanol production. The reported exergy efficiencies were 87.5% for pre-treatment, 65% for pentose fermentation, 48% for SSF and 68.2% for purification (distillation and dehydration). Moreover, these authors performed a comparison via exergy analysis of SHF and SSCF pathways (Ojeda et al., 2011a).

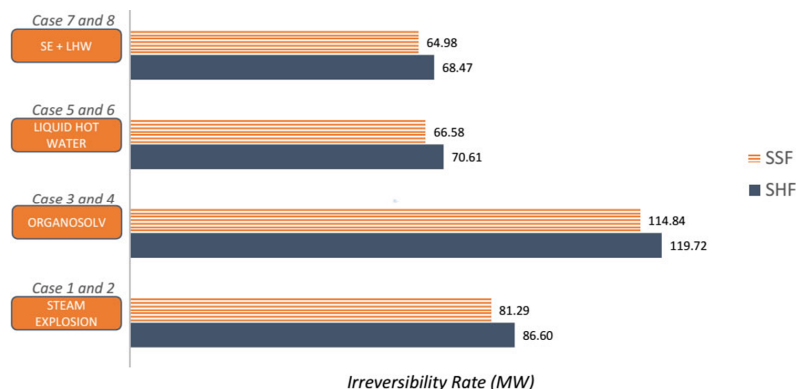
Palacios-Bereche et al. carried out an exergy analysis and an exergy cost analysis of the bioethanol production by enzymatic hydrolysis of sugarcane bagasse integrated with and without the conventional process. These authors, adopted steam explosion (case 1) as the pre-treatment technology, and found an exergy efficiency of 63% for the enzymatic hydrolysis process using an evaporation system, and considering 10% of solids content in the hydrolysis reactor (Palacios-Bereche et al., 2013).

Hence, the results of this study are in the lines of earlier literature reported for exergy efficiency of cases 1, 2 and 4. On the basis of the results, a performance comparison of the SHF and SSF configurations in terms of exergy efficiency and irreversibility rate are summarised in Figures 9 and 10, respectively.

**Figure 9** Exergy efficiency of the SHF and SSF routes (see online version for colours)



**Figure 10** Irreversibility rate of the SHF and SSF routes (see online version for colours)



## 5 Conclusions

The biological conversion of sugars into ethanol remains a critical step (process integration) in first- and second-generation ethanol processes. As a result, ethanol production from second-generation feedstocks shows significant potential for technical improvements. Consequently, to reach an economically viable ethanol production using bio-based materials, each step (pre-treatment, hydrolysis and fermentation) has to be optimised in order to obtain high ethanol rate/yields with the lowest possible cost.

Thus, in the present study, two different configurations for second-generation ethanol production from sugarcane bagasse focusing on wine production were simulated and analysed combined with pre-treated bagasse obtained from four pre-treatment biomass techniques. On the basis of the exergy efficiency performed, the SSF configurations show the highest values due to the increased rate of saccharification compared with those obtained in SHF processes.

In terms of total wine productivity, it is evident that wine production for the cases studied is a function of the inlet mass flow rate of pre-treated sugarcane bagasse. The best alternative to improve wine production processes via enzymatic technology, SHF and SSF configurations, were obtained through cases 3 and 4, increasing the wine production when compared with the other technologies by 10.94% and 10.86%, respectively. Furthermore, when considering the use of xylose liquor (pentose) regarding the production of biogas, cases 1 and 2 represent the best technological options.

Moreover, a sensitivity analysis varying the dosage of the enzymes showed that it does not represent a significant fraction (<2%) in increasing the total wine production for all cases. However, a detailed application of techno-economic analysis in this matter is necessary to estimate their impact on second-generation bioethanol production.

## Acknowledgements

The authors acknowledge FAPESP (São Paulo Research Foundation) for grant 2012/23049-2, and CNPq (Brazilian National Council of Scientific and Technological Development) for grant 306033/2012-7.

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**Nomenclature**


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1G	First-generation bioethanol production
2G	Second-generation bioethanol production
b	Specific exergy, kJ/kg
$B_{\text{tot}}$	Exergy rate, kW
CBP	Consolidated bioprocessing
FPU	Filter paper unit
H <sub>2</sub> O-WASH	Water for pre-treated bagasse washing
H <sub>2</sub> O-HYD	Water for enzymatic hydrolysis of bagasse
JUICEVAP	Vapour of 5th effect-glucose liquor concentration system
PEN-LIQ	Pentose liquor
SHF	Sequential hydrolysis and fermentation
SSCF	Simultaneous saccharification and co-fermentation
SSF	Simultaneous saccharification and fermentation
STEAM-EV	Steam evaporation system
t/h	Tonne per hour
w/v	Weight per volume, %
w/w	Weight by weight, %

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