Sugarcane molasses-based biorefinery: Organic acids and ethanol production

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Abstract: Sugarcane molasses are the largest produced waste in sugar mills; in the last harvesting cycle 2,178,131 tons were obtained and only 2.46% were used for transformation processes. Molasses has great potential to be the main feedstock in a biorefinery concept. Its composition rich in fermentable sugars and its availability are interesting features considered in this study. Through the Aspen Plus© software a multi-product biorefinery scenario was design and analyzed, technically and economically. The three main products considered were ethanol, lactic acid and succinic acid. The overall process consists of an initial stage of hydrolysis or inversion of the sucrose present in the molasses to reducing sugars followed by a specific dilution for fermentation of each of the products considered, as well as subsequent separation and purification operations. Plant efficiencies conversions were 3.24 kg of molasses/L of ethanol, 3.08 kg of molasses/kg of lactic acid and 9.25 kg of molasses/kg of succinic acid. The economic assessment was positive for organic acids production. Ethanol production had a slightly worst economic performance compared to the other processes, but the proposed scenario managed to obtain a profitability index of 1.02. The expense recovery ratio of the whole biorefinery was 1.35 which means a surplus of 35% after the project investment has paid for itself. The biorefinery's robustness in the economic aspect comes from organic acids production; meanwhile, the social and environmental impacts are from ethanol production.

Keywords: Sugarcane molasses; Biorefinery; ethanol; lactic acid; succinic acid

Introduction

The sugarcane industry in Mexico has suffered a lag in its competitiveness, due to diverse and complex factors, such as: low productivity at field and in sugar mills production, unusual fluctuations in sugar prices (domestic and international prices), consumption substitution of sucrose for high fructose syrups from corn and artificial sweeteners, high production costs, inefficient processes, no diversification of the products in the sugar mills across the country, and non-clear use of the by-products generated in the sugarcane processing (Aguilar et al. 2012)

The 87.5% of sugar produced in the world is from sugarcane, hence its relevance as the third crop more cultivated worldwide, after other cereals such as corn, rice and wheat. Mexico is the sixth major producer of sugarcane; the other five countries ahead of Mexico in sugarcane production are Thailand, Pakistan, China, India and Brazil (FAOSTAT Database 2021). In the 2021-2022 harvesting cycle of sugarcane, Mexico had a cultivated surface of 799,774 hectares; 54,680,830 tons of sugarcane was harvested and 6,185,050 tons of sugar was produced in 15 sugarcane producer states (see Figure 1) (CONADESUCA Database 2022). While Mexico has managed to position itself as a major producer of sugar in the world, the cultivation and processing of sugarcane occurs under hostile conditions. The conditions such as socioeconomic, environmental and technical aspects create a complex problem to be solved in order to improve the competitiveness of this agribusiness (Sentíes-Herrera et al. 2014).

The aforementioned problematic is to be attended by the following points:

- Development and improvement of the human force in the sugar mills.
- Sugar mills modernization.
- Generation of sustainable processes.
- Transformation of sugar mills into biorefineries.
- Diversification of the production chain. (Aguilar-Rivera et al. 2012)

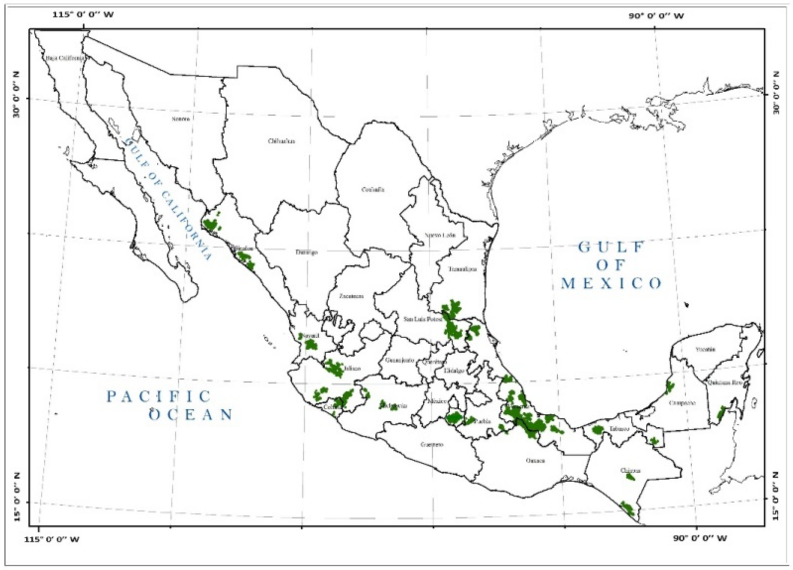

Figure 1. Mexican sugarcane mills distribution

The sugarcane agro-industry generates a great number of by-products in different processing lines. One of the main by-products is molasses, also known as black treacle.

Molasses is a very viscous liquid; its color is usually dark brown, almost black. The composition of molasses is very rich in reducing sugars, sucrose, vitamins, fats, minerals and other components. In Mexico, usage of molasses is mostly directed for alcohol production and cattle feed (SAGARPA Database 2016). Molasses are an interesting feedstock with great potential of transformation in high-value products: high carbohydrates concentration and a low cost feedstock (Gomes et al. 2017). Compared to lignocellulosic materials, molasses doesn't need sophisticated pretreatments (van der Merwe 2010), also, molasses disposition does not interfere with the production of food crops (Tan et al. 2008). Nonetheless, the availability of molasses is strongly linked to sugarcane production; as well, the composition of molasses could contain contaminants, ashes, that directly affect in fermentation systems (Cardona, C. A., Sanchez, O. J., Gutierrez 2010).

In agreement with the statistical report of the National Committee for the Sustainable Development of Sugarcane (CONADESUCA) in the latest harvesting cycle (2021-2022) 2,178,131 tons of molasses were obtained. The historical average annual production of molasses from 2012 until 2022 is 2,050,810 tons. From this production, only 2.46% of molasses is used for alcohol food grade production. Therefore, it is assumed that in Mexico there is availability of this by-product. The main focus is to set a biorefinery concept around the exploitation of by-products in the sugarcane industry (de Jong and Jungmeier 2015).

Nowadays, molasses has been object of study for the production of biofuels and bioproducts; ethanol, butanol, hydrogen, acetone, hydrogen, methane, butanediol, lactic acid, succinic acid, among others through fermentation processes (Fernández-López et al. 2012; Rendon-Sagardi et al. 2014; Castañeda-Ayarza and Cortez 2017; Kumar et al. 2018; Merwe et al. 2013; Gao et al. 2022; Kingsly et al. 2022; Vilela et al. 2021; Tinoco et al. 2121; Zhang et al. 2020; Sun et al. 2019; Chan et al. 2012). Sugarcane and beet molasses are also considered as an attractive substrate for catalytic processes for the production of hydroxymethylfurfural through degradation of molasses and other water soluble sugars (Gomes et al. 2017). The implementation of sugarcane biorefineries for the production of ethanol, lactic acid and electricity takes into consideration at some point the utilization of molasses, but the main feedstock is lignocellulosic materials such as sugarcane bagasse (Mandegari et al. 2017a).

The two main objectives a biorefinery must accomplish are: the displacement of fossil fuels in favor of fuel utilization that comes from renewable resources; this is known as *energetic goal*. Besides, the economic goal of biorefinery must be met through the establishment of a robust bioproduct industry (Bozell and Petersen 2010).

In recent years, some organic acids like lactic acid and succinic acid have become an important focus due to their great versatility as building blocks; i.e., their flexibility to be marketed just as they are or to be transformed in other products known as high specialty chemicals (Werpy and Petersen 2004; Borodina and Nielsen 2014).

Molasses from sugarcane has many special characteristics to be considered as an ideal feedstock in a biorefinery concept. The main goal of this work is to present a new proposal for the sugar mills for the diversification of its product offering. We present a biorefinery scenario with molasses as the central feedstock for the multiproduct generation of organic acids and ethanol. To our knowledge, there are no proposals of biorefineries scenarios with molasses from sugarcane as the main sugar platform.

Materials and Methods

The simulation procedure of the organic acids and ethanol biorefinery was carried out in Aspen Plus© v. 8.8. The required raw material, molasses, was 150 ton/day. In Table 1, the chosen flow distribution is shown for each process. In Figure 2 it is shown the general process diagram followed for the biorefinery simulation.

Table 1. Amount of molasses for each process of biorefinery

Biorefinery process	Molasses (ton/day)
Ethanol	100
Lactic Acid	25
Succinic Acid	25
Total	150

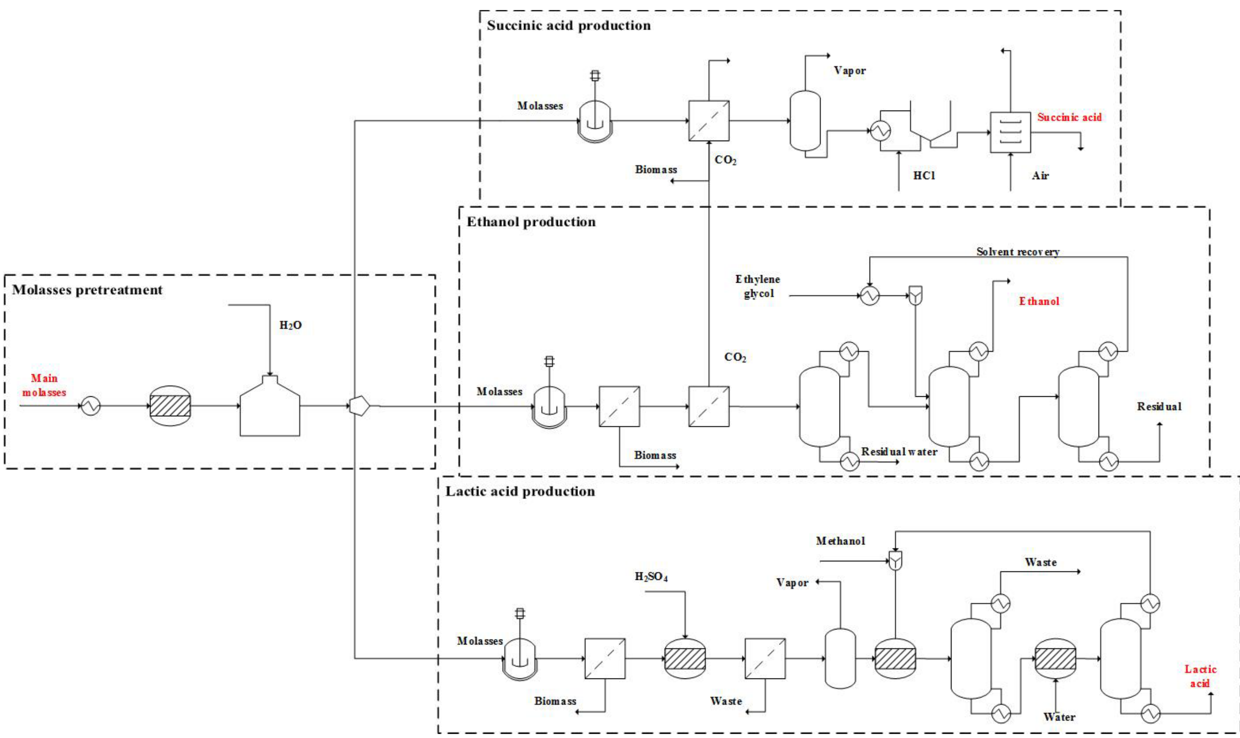

Figure 2. Proposed biorefinery scheme for the simulation in Aspen Plus

The amount of molasses designated for each simulation process corresponds to the production in Michoacan sugar mills. Michoacan has three operational sugar mills, which generated 40,332 tons of molasses in the 2021-2022 sugarcane harvesting cycle, (CONADESUCA Database 2021). The non-random two liquid (NRTL) thermodynamic model is applied to calculate the activity coefficients of the liquid phase and the Hayden-O'Connell equation of state was used to model the vapor phase (Moncada et al. 2013).

For the fermentation steps of the three processes, it was assumed an Arrhenius type kinetic model for bioprocesses (Arni et al. 1999; Brandam et al. 2008). Equation 1 shows the dependence of specific growth velocity with respect of temperature:

$$\mu = Ae^{-Ea/RT} \tag{1}$$

The kinetic parameters required for the simulation procedure are shown in Table 2 (Hujanen and Linko 1996; Corona-González et al. 2008; Ortiz-Muñiz et al. 2010).

Table 2. Kinetic parameters for simulation purposes in the fermentation stage of the biorefinery

Microorganism	**Fermentation stage**	**Activation energy (kcal/mol)**	**Pre-exponential factor**	**Fermentation temperature (°C)**
Saccharomyces cerevisiae	Ethanol	15.6	1.05×10^9	30
Lactobacillus casei	Lactic acid	19.48	1.38×10^9	37
Actinobacillus succinogenes	Succinic acid	19.48	1.38×10^9	37

The economic analysis for the proposed biorefinery was performed by Aspen Process Economic Analyzer (APEA). The economic parameters considered for this analysis are typical for the Mexican case; estimations were calculated in USD dollars. It was chosen a 20-year period with a 40% annual income tax. It was considered the straight line method for capital depreciation.

Molasses price were considered according to local markets (Secretaria de Economia Database 2017). Supply prices for the biorefinery are in agreement with Mexican conditions. Utilities costs for electricity, steam, water, costs for supervisors and operators are according to Mexican conditions (Vargas Santillán et al. 2016).

Equipment calculations, capital costs of process units, operating costs, among other necessary data and correlations were obtained from APEA (Peters et al. 1991). Other prices considered for the economic analysis are summarized in Table 3 (ALIBABA 2017; ICIS 2017).

Table 3. Prices considered for supplies and products in the biorefinery

Item	**Price**	**Unit**
Molasses	150	USD/ton
Microorganisms	0.03	USD/kg
Ethylene Glycol	1.43	USD/kg
pH controller	0.08	USD/kg
H_2SO_4	0.027	USD/kg
Methanol	0.45	USD/kg
HCl	0.027	USD/kg
Ethanol	0.7	USD/L
Lactic acid	2500	USD/ton
Succinic acid	3000	USD/ton

Pretreatment Stage

The pretreatment of molasses is basically composed of two steps: hydrolysis of the sucrose in molasses in order to obtain 6-carbon sugars. The next step is dilution of molasses concentration, specific for each fermentation stage. Molasses composition is variable; therefore, for simulation purposes, it assumed that its composition is constant throughout the whole process. Composition of molasses is shown in Table 4 (Merwe et al. 2013).

Table 4. Composition assumed for molasses for the simulation procedure

Component	Percentage (%w/w)
Water	37.7
Solids	
Sucrose	25.2
Reducing sugars	37.1

Hydrolysis conditions are 60 °C and 1 atm. It was assumed all sucrose is converted in hexoses (Jacques et al. 2003). Dilutions were simulated in agitated tanks to guarantee the desired concentration for the next stage.

Lactic acid production

Lactic fermentation required a maximum molasses concentration of 150 g/L. This process was simulated in a continuous reactor at standard pressure conditions and at temperature of 37 °C. It was assumed that this conditions are ideal for *Lactobacillus casei*; the chosen microorganism for lactic acid production (Thakur et al. 2017). Purification of lactic acid was performed through an esterification, hydrolysis and a distillation system. These are typically steps used for lactic acid separation from the aqueous phase present in the broth.

Succinic acid production

The maximum molasses concentration needed for the production of succinic acid is 150 g/L. *Actinobacillus succinogenes* was the considered microorganism. A continuous reactor was also used in the simulation at 37 °C of temperature and 1 atm of pressure. Purification of succinic acid was performed through two main steps: evaporation of other organic acids presents in the broth mix and the subsequent crystallization of succinic acid solution.

Ethanol production

Saccharomyces cerevisiae is the yeast considered for alcoholic fermentation. The concentration of molasses was 150 g/L in a continuous reactor. It was assumed that molasses contained enough nutrients for this step and only a pH controller was added. The concentration of the broth and the dehydration of the water-ethanol solution was simulated as an extractive distillation process with ethylene glycol as a solvent.

Results and Discussion

The overall material balance is shown in Table 5. The fermentation stages showed a molasses transformation yield of 49% for ethanol, 99% for lactic acid and 63% for succinic acid.

Table 5. Materials balance for each product in the biorefinery

Biorefinery Process	Input: Molasses (ton/day)	Output: Product (ton/day)
Ethanol	100	22.63
Lactic Acid	25	8.1
Succinic Acid	25	2.7

In order to produce a liter of ethanol 3.24 kg of molasses is required, the plant efficiency conversion (PEC) reported is 4.98 kg of molasses per liter of ethanol. Basically, the proposed scenario for ethanol production is working as expected and has yields similar to the literature (Silalertruksa et al. 2017).

For the organic acids production, lactic acid has a yield or a plant efficiency conversion (PEC) of 3.08 kg of molasses/kg of lactic acid. This conversion is slightly inferior to the ones reported for lignocellulosic materials ; 2.02 kg lignocellulosic material/kg of lactic acid (Mandegari et al. 2018). On the other hand, the plant efficiency for succinic acid is low compared to the other two biorefinery processes; for every 9.25 kg of molasses converted one kg of succinic acid is obtained.

The energy requirements of each biorefinery process are shown in Figure 3.

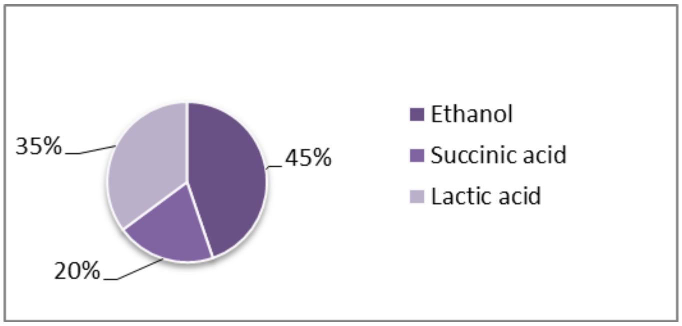

Figure 3. Energy requirements for each process in the biorefinery

According to the energy requirements reported, ethanol requires the greatest amount of energy for its production, followed by lactic acid also requires an important amount of energy due to the many steps in its purification stage. Succinic acid only requires the 20.13% of the whole energy required by the biorefinery. As we know, energetic demands for ethanol production are similar to those reported in the literature (Rincón et al. 2014).

Analysis of the economic performance of the biorefinery

The evaluation of the economic performance of the biorefinery was carried out in APEA. Various aspects such as operating, raw materials, utilities, labor, maintenance and general plant costs were calculated through this tool integrated in the Aspen Plus software. In Figure 4, it is shown the share of the total production cost for each process in the biorefinery.

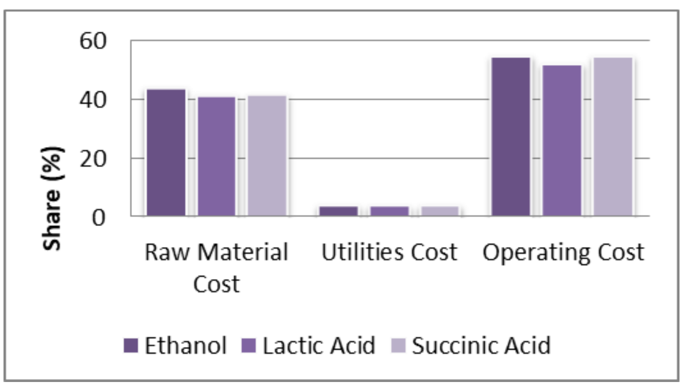

Figure 4. Share of total production costs components.

In this kind of project, the raw material cost often takes up to 50% of the total operating cost. In the proposed biorefinery scenario for Mexican sugarcane mills, molasses are an affordable feedstock that represents up to 44% of the total operating cost (Quintero et al. 2013). Although the operation costs take up to 50% of the total production costs, this effect is due to many stages required in purification of ethanol and lactic acid purification.

In Figure 5 it is shown the comparison between total production cost and total product sales. This is an integrated approach of the biorefinery. The product sales generated mostly by lactic acid production and succinic acid counteract positively the total production costs.

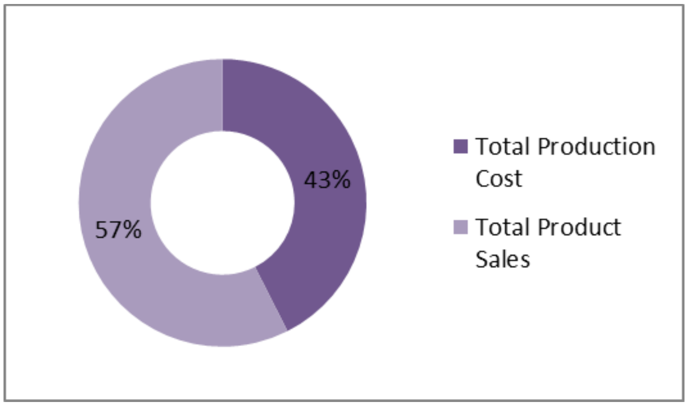

Figure 5. Comparison between total production cost and total product sales

There are several economic benchmarks to determine if a project is suitable for its implementation. Basically, these indicators measure the profitability of potential investments. In this work, it was taken in account the internal rate of return (IRR), the profitability index (PI) and the expense recovery ratio (ERR) in order to evaluate the viability of the proposed biorefinery. In Table 6 it the results from APEA are presented for each process. The biorefinery was analyzed this way in order to show the impact of every process on the economic aspect.

Table 6 Results from APEA for each process analyzed

Production Process	Desired IRR (%)	IRR (%)	Profitability Index (PI)
Ethanol	20	21.29	1.02
Lactic acid	20	26.68	1.12
Succinic acid	20	23.07	1.05

The prices considered for the final products and for the above analysis were 0.7 USD/L for ethanol, 2.5 USD/kg for lactic acid and 3.0 USD/kg for succinic acid. These are minimum prices in the global market. Even though the IRR obtained for each process is slightly above in comparison with the desired IRR, the biorefinery scenario seems plausible. Also, when the profitability index of an investment project is greater than one, it is advisable that the project be accepted; in the previous analysis, every process has a PI greater than one.

The expense recovery ratio is an economic benchmark that shows very easily how an investment has regained its costs. It is often calculated as the relation between the total revenue of an investment by its total expenses. In this work, the ERR was evaluated for the whole biorefinery showing a positive ERR of 1.35. This result could be interpreted as sign of surplus of the whole investment project.

Conclusions

A biorefinery scenario bio-based on molasses simulated in Aspen plus was studied due to lack of proposals where molasses is the main feedstock in a biorefinery. For the main fermentation stages; that are the 'bottle neck' of the biorefinery, it was reached maximum conversion yields of sugar into the desired products. Ethanol and lactic acid production are the most energy demanding processes, but overall, they are the most profitable products with 85% of total product sales. In the other hand, succinic acid has a low energy demand and only accounts for 15% of total product sales, however this product is the most expensive and its production could be subject of optimization.

The plant efficiencies conversions of ethanol and lactic acid are in accordance with those found in the literature. Succinic acid has a lower efficiency, as previously mentioned, but it is the most promising product in the biorefinery with positive market projections in the future.

The configuration proposed in Aspen Plus shows positive results in the economic parameters; IRR, PI and ERR. Also, it can be seen that organic acid production contributes to the economic aspect of the biorefinery. Meanwhile, ethanol production contributes to the environmental field of the biorefinery. The whole biorefinery contributes to social aspects; such as: generations of jobs, reactivation of economic activities in rural zones and improvement of quality life for those involved in these kind of projects.

It is recommended to modify the plant configuration in order to optimize the production of high value products: consider novelty purification processes, adjust the amount of molasses designated for each product and regard cogeneration systems to improve energy consumption.

Abbreviations: APEA: aspen process economic analyzer; ERR: expense recovery ratio; IP: profitability index; IRR: internal rate of return.

Author contributions: T.M-R: performed the simulation, data generation and wrote the manuscript; A.J. C-M: conceptualization and design the process; N. A-R: provide data of raw materials; All authors: analysis and interpretation of results, editing the final manuscript

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