Techno-economic and Environmental Assessment of an Olive Stone based Biorefinery

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Abstract

Olive tree cultivation is spreading worldwide as a consequence of beneficial effects of olive oil consumption. Olive oil production process and table olive industries are the major

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sources of olive stones. Currently, this by-product is used in direct combustion to produce energy as electricity or heat. However, there are other possibilities for taking full advantage of a renewable source of interesting compounds. In this work the techno-economic and environmental assessment of two biorefinery schemes and its comparison with the direct combustion (base case) of this residue are presented. The first biorefinery scheme describes the integrated production of xylitol, furfural, ethanol and poly-3-hydroxybutyrate (PHB). The second biorefinery scheme considers the production of xylitol, furfural, ethanol and PHB integrated to a cogeneration system for producing bioenergy from the solid residues resulting from the mentioned processes. The results showed that in the first biorefinery scheme, the net profit margin is approximately 53%, while the second present a net profit margin of 6%.

Keywords: Biorefinery, olive stone, ethanol, xylitol, PHB, furfural

1. Introduction

Currently, most energy and chemicals are derived from fossil raw materials, although there are several environmental, economic and social concerns related to their extraction and use. These facts have increased the interest in the use of renewable raw materials. The replacement of the fossil raw materials either fully or partially is an objective in many countries, including Spain. Since the fossil supply in this country is almost entirely of foreign origin, is of special interest the use of local biomass such as agricultural, forest, agro-industrial and industrial wastes, due to its low cost and large availability.

Because of the reported beneficial effects of olive oil consumption, the olive tree cultivation has been propagated worldwide and it is nowadays present in countries as diverse as the United States, Argentina or Australia. Currently, this crop presents a global cultivated area of almost 10 million hectares with an annual production of approximately 18 million tons of olives (FAOSTAT, 2014). Olive oil and pitted table olive production are the most important agrifood industries in the Mediterranean countries, with Spain being the largest producer in the world. One important by-product generated in olive oil extraction and pitted table olive production is olive stone. Olive stone represents 10-30 % wt (Garrido-Fernández et al. , 1997) of the fruit, which implies an annual production of approximately three million tons. For instance, in the 2009/2010 season, the olive oil and table olives world production were 2.97 and 2.37 million tons, respectively (The International Olive Council, 2012). In the same season, approximately 0.17 million tons and 2.1 million tons of stone from table olives and olive oil industries, respectively, were produced. Currently, the main use of this by-product is the direct combustion to produce energy as electricity or heat (Romero-García et al. , 2014).

Apart from its use as raw material for producing heat and electricity, crushed olive stones have also been considered as raw material for other kind of value-added products. For instance, olive stones contain extracts with high antioxidant capacity (approximately 5.5% dry basis), containing mainly hydroxytyrosol and tyrosol (Fernández-Bolaños et al. , 1998). These antioxidants have application in the food, cosmetic, functional food and nutraceuticals industries (Spizzirri et al. , 2011). Hydroxytyrosol is able to inhibit or retard the rate of growth of a broad range of bacteria and fungi (Elbir et al. , 2012). The extraction of phenolic compounds offers a double advantage. Firstly the recovery of the bioactive products with antioxidant capacity and high value-added, improves the economic viability of the process. Besides, the toxicity of the subsequent pre-hydrolyzed can be reduced, increasing the yield of the enzymatic hydrolysis or fermentation steps. On the other hand, olive stone is a lignocellulosic material with reducing sugar and lignin content of approximately 50% and 40%, respectively. This reducing sugar fraction allows obtaining different value-added products. For instance, in this work four products are considered: Ethanol and furfural, because of being renewable fuels; xylitol, because of its application in the nutraceutical industry; and PHB in order to evaluate the production of an alternative polymer.

In this work the direct combustion of the olive stone (base case) and two biorefinery scenarios are techno-economic and environmentally assessed. The first biorefinery scenario presents the integrated production of xylitol, furfural, ethanol and poly-3-hydroxybutyrate (PHB). In the second scenario, the additional production of bioenergy from the solid residues integrated to de production of xylitol, furfural, ethanol and PHB is considered.

2. Materials and Methods

2.1. Raw material

Olive stones were supplied by the olive-oil mill factory “S. C. A. Unión Oleícola Cambil” located in Jaén, Spain. The stones were separately removed from the olive pomace with an industrial pitting machine, with a 6 mm sieve separator, which is the standard size in this industrial process, soaked in water, washed to free them from any adhering flesh, air-dried and then dried for 24 h at 50 °C. The composition of the raw material was determined according to NREL (National Renewable Energy Laboratory, Golden, CO, USA) analytical methods for biomass.

2.2.Scenarios description

In this work a base case and two biorefinery scenarios were techno-economic and environmentally assessed. The base case considers the current use for this raw material: power production through a direct combustion process. In this scheme, the olive stone is submitted to a combustion process at 850 °C and 60 bar. The resulting stream goes through a gas turbine where the pressure decreases up to 1 bar to produce electricity. The first biorefinery scheme for producing xylitol, furfural, ethanol and PHB is presented in Fig. 1. In this process, the raw material is submitted to pretreatment and hydrolysis, where the xylose and glucose rich fractions were extracted. Then the xylose-rich fraction was destined to produce xylitol (80%) and furfural (20%). On the other hand, the glucose-rich fraction was used to produce ethanol (80%) and PHB (20%). Finally, in the second biorefinery scheme, the solid residues resulting from xylitol, furfural, ethanol and PHB processes, which are lignin and biomass, are used to produce power and heat through a cogeneration system (See Fig. 2).

Figure 1.

Figure 2.

2.3.Simulation Process

For the base case as well as for the two biorefinery scehmes, flowsheet synthesis was carried out using process simulation tools. The objective of this procedure was to generate the mass and energy balances for calculating the raw materials, consumables, utilities and energy requirements. The main simulation tool used is the commercial package Aspen Plus v8.0 (Aspen Technology, Inc., USA). Matlab was also used to perform mathematical calculations especially for kinetic analysis. The kinetic models used for calculations of hydrolysis steps were reported by Jin et al. and Rinaldi & Schüth (Jin et al. , 2011, Rinaldi and Schüth, 2009). Kinetic model for detoxification were reported by Martinez et al. (Martinez et al. , 2001). The fermentation stage for fuel ethanol production was calculated using the kinetic model reported by Leksawasdi et al. (Leksawasdi et al. , 2001). The kinetic model used for the calculation of PHB production was reported by Shahhosseini (Shahhosseini, 2004). For xylitol, the fermentation conditions were adapted from (Tochampa et al. , 2005). For furfural production, the reaction conditions were taken from Agirrezabal-Telleria et al. (Agirrezabal-Telleria et al. , 2013). The Non-Random Two-Liquid (NRTL) thermodynamic model was applied to calculate the activity coefficients of the liquid phase and the Hayden-O'Conell equation of state was used for the description of the vapor phase. Also the UNIFAC-DORTMUND and Soave Redlick Kwong models for liquid and vapor phases were needed when the NRTL model do not predict properties (e.g. Liquid-Liquid separations and distillation columns).

2.3.1. Sugar extraction

The olive stone is initially submitted to a process consisting of two hydrolysis steps in order to achieve the sugar extraction. In the first stage the hemicellulose fraction is hydrolyzed with sulfuric acid (2.4% by weight) at a temperature of 100 °C. The result of this hydrolysis is a non-converted solid fraction and the rich-pentose liquor. This resulting stream is separated by filtration. Then the solid fraction, rich in cellulose and lignin is submitted to a second acid hydrolysis step with sulfuric acid (1.2% by weight) at 2.68 atm and 150 °C to obtain the rich-hexoses liquor and a solid residue rich in lignin. The resulting lignin is then sent to the cogeneration system, for the second scenario.

As a result of the decomposition reactions of sugars during the two hydrolysis steps, furfural and hydroxymethyl furfural (HMF) are obtained. Then, detoxification technology is applied and it is based on overliming (Martinez, Rodriguez, 2001, Millati et al. , 2002, Purwadi et al. , 2004). This procedure is carried out to avoid poisoning and inhibition by the acids, furfural and HMF in the fermentation stages.

The resulting xylose-rich liqour is divided into two fractions. The 80% is submitted to the xylitol production process and the other 20% is used to produce furfural. In the case of the glucose-rich liquor, it is used to obtain ethanol and PHB.

2.3.2. Xylitol production

Xylitol is synthesized from xylose by the yeast *Candida mogii*. Initially the liquor is sent to a sterilization process at 121 °C in which the biologic activity is neutralized. Once the culture media is sterilized the fermentation with *C. mogii* is performed according to the kinetic expression reported by Tochampa et al. (Tochampa, Sirisansaneeyakul, 2005), at 30 °C under aerobic conditions (dissolved oxygen concentration of 20%). After fermentation, the resulting stream is filtered to separate the biomass and the temperature is increased at 40 ºC and a flash operation is used to concentrate the obtained xylitol. The next step for isolating the metabolite from the fermentation broth consists on an evaporation to eliminate the excess of water for facilitating the concentration by crystallization, adding ethanol in order to decrease drastically the xylitol solubility and supersaturate the solution to carry out the crystallization at 5 °C (Parajó et al. , 1998).

2.3.3. Furfural production

Furfural is obtained from the xylose-rich liquor via xylose cyclodehydration using air as stripping agent for removing the product while is produced (Agirrezabal-Telleria, Gandarias, 2013). First the liquor is sent to a reactor at 180 °C and 10 bar. Then, air is feeding into the reactor at a ratio of 30:1 air to feed. The resulting stream is then depressurized to recover the liquid fraction. After, the mixture is sent to a liquid-liquid extraction process with toluene as solvent with 1:1 v/v ratio to recover the furfural from the water. Finally, the solvent-furfural stream is submitted to a distillation process where the furfural is obtained as bottom product.

2.3.4. Ethanol production

Initially the liquor is sent to a sterilization process at 121 °C in which the biologic activity is neutralized. Later the fermentation process is carried out based on the kinetic expressions reported by Leksawasdi et al. (Leksawasdi, Joachimsthal, 2001), at 30 °C using *Zymomonas mobilis* as microorganism. Afterwards, the cell biomass is separated from the culture broth.

After the fermentation stage, the culture broth containing approximately 7-10% (wt/wt) of ethanol is taken to the separation zone, which consists of two distillation columns. In the first column, ethanol is concentrated nearly to 50-55% by weight. In the second column, the liquor is concentrated until the azeotropic point (96% wt) to be led to the dehydration zone with molecular sieves for obtaining an ethanol concentration of 99.7% by weight (Pitt Jr et al. , 1983).

2.3.5. Poly-3-hydroxybutyrate production

Firstly the hexose-rich liquor is submitted to a sterilization stage of the culture broth also involving a nitrogen source ($(NH_4)_2SO_4$), at 122 °C and a pressure of 10 bar. The ratio of the nitrogen source to the carbon source is 0.16 g/g. Then, the culture with an appropriate glucose concentration (approximately 8% by weight) undergoes fermentation. The glucose fermentation is performed with *Ralstonia etropha* as microorganism according to Shahhosseini (Shahhosseini, 2004), at 30 °C in presence of $(NH_4)_2SO_4$ as nitrogen source. Then, a digestion which consists of cell lysis with chemical agents such as sodium hypochlorite assisted by temperature (Herron et al. , 1978, Moncada et al. , 2013) is done. Once the biopolymer is extracted, residual biomass is separated by centrifugation. The resulting solution after centrifugation is washed in order to remove impurities to finally remove water by evaporation and spray drying to obtain PHB approximately at 98% (wt/wt).

2.3.6. Energy cogeneration

For this section the technology used for cogeneration is the biomass integrated gasification combined cycle (BIGCC), as shown in Fig. 2 (Balat et al. , 2009, Herron, King, 1978, Rincón et al. , 2013). Basic elements of BIGCC system include biomass dryer, gasification chamber, gas turbine and heat steam recovery generator (HRSG). Gasification is a thermo-chemical conversion technology of carbonaceous materials (coal, petroleum coke and biomass), to produce a mixture of gaseous products (CO, CO_2, H_2O, H_2, CH_4) known as syngas added to small amounts of char and ash. Gasification temperatures range between 875 and 1275 K (Ahmed and Gupta, 2012). The gas properties and composition of syngas changes according to the gasifying agent used (air, steam, steam-oxygen, oxygen-enriched air), gasification process and biomass properties (Ahmed and Gupta, 2012). Syngas is useful for a broader range of applications, including direct burning to produce heat and power or high quality fuels production or chemical products such as methanol (Rincón et al. , 2014, Xu et al. , 2011). A gas turbine is a rotator engine that extracts energy from a flow combustion gas. It is able to produce power with an acceptable electrical efficiency, low emission and high reliability. The gas turbine is composed by three main sections: compression (air pressure is increased, aimed to improve combustion efficiency), combustion (adiabatic reaction of air and fuel to convert chemical energy to heat) and expansion (obtained pressurized hot gas at high speed passing through a turbine generating mechanical work) (Najjar, 2000). The HRSG is a high efficiency steam boiler that uses hot gases from a gas turbine or engine to generate steam, in a thermodynamic Rankine Cycle. This system is able to generate steam at different pressure levels. According to process requirements a HSRG system can use single, double or even triple pressure levels.

2.4.Energy consumption

The estimation of the energy consumption was performed based on the results of the mass and energy balances obtained from the simulation. Then, the required thermal energy in the heat exchangers and re-boilers was calculated, as well as the electric energy needs for the pumps, compressors, mills and other equipment.

2.5.Economic assessment

The capital and operating costs were calculated using the software Aspen Economic Analyzer V8.0 (Aspen Technologies, Inc., USA). On the other hand specific parameters regarding to Spain conditions such as the raw material costs, income tax (35%), annual interest rate (6%) and labor salaries, among others, were incorporated in order to calculate the production costs per unit for the different obtained products. This analysis was estimated in US dollars for a 10-year period. The Capital depreciation was calculated using the straight-line method. As a result of this analysis, the biorefinery total cost is expressed by the sum of the following items: Capital depreciation, raw material costs, utilities costs and operating costs. Besides, the net and unit profit margin of the biorefinery and its products, respectively, as well as the unit production cost are shown as parameters for performing the economic assessment.

2.6.Environmental assessment

The Waste Reduction Algorithm WAR, developed by the National Risk Management Research Laboratory from the U.S. Environmental Protection Agency (EPA) is used as the method for the calculation of the Potential Environmental Impact (PEI). The PEI for a given mass or energy quantity could be defined as the effect that those (energy and mass) will have on the environment if they are arbitrary discharged. The environmental impact is a quantity that cannot be directly measured; however, it can be calculated from different measurable indicators. The WAR GUI software incorporates eight categories: Human toxicity by ingestion (HTPI), human toxicity by dermal exposition or inhalation (HTPE), aquatic toxicity potential (ATP), Global warming (GWP), Ozone depletion potential (ODP), Photochemical oxidation potential (PCOP) and acidification Potential (AP). This tool considers the impact by mass effluents and the impact by energy requirements of a chemical process, based on the energy and mass balances generated in Aspen Plus. Then the weighted sum of all impacts ends into the final impact per kg of products.

3. Results and discussion

3.1.Raw material

Olive stone was characterized in terms of ash, extract, acetyl groups, moisture content and lignocellulosic composition (representing xylose more than the 80%) and lignin (See Table 1). In addition olive stone contains pulp (1.6%) and skin (1.4%) residues.

Table 1.

3.2.Process Simulation

For simulation purposes a raw material flow of 10 ton/h (approximately 80,000 ton/year) is considered. According to the Ministerio de Agricultura, Alimentación y Medio Ambiente of Spain, approximately 6 million tons of olive are produced per year in Spain and the 35% of this production is concentrated in Jaen. This means that approximately 210,000 to 630,000 tons of olive stone are produced in this Province. This means that the annual feedstock requirement corresponds approximately to the 4.4%-13% and the 13-38% of the total availability of the olive stone in Spain and the Province of Jaen, respectively. Simulations of the studied scenarios were used to generate their respective mass and energy balance sheets, which are the basic input for the techno-economic and environmental assessments. Results are shown in Table 2. These evaluations were carried out considering that for the first scenario, all the raw material is destined to electricity production through combustion. On the other hand, for scenarios 2 and 3 the feedstock distribution is as follows: 80% of the hexose-rich liquor for producing ethanol and 20% for PHB, 80% of the pentose-rich liquor for obtaining xylitol and 20% for furfural production.

Table 2.

3.3.Economic Assessment

Distribution and technologies included in each scenario directly affect production costs. In this sense, the economic analysis is focused on two parameters: production cost and net profit margin. The annualized costs for each scenario are presented in Table 3. As can be seen the feature that contributes with a higher proportion on the total costs are the operating costs followed by utilities, raw materials and depreciation. Moreover, scenario 2 shows higher values due to the addition of the cogeneration scheme. This addition represented an increment of the 14% on the annualized costs based on the second scenario. Comparing to the base case, utilities for both biorefinery schemes are higher. Besides, the total annualized costs for scenarios 2 and 3 show an increment of 23.3% and 33.7%, respectively, with respect to the base case.

Table 3.

In Table 4, the unit cost for each product is presented. Besides, in Fig. 3 the profit margin for each product in both biorefinery schemes as well as the net profit margin is presented. For the given scheme, furfural and ethanol are not profitable. However, xylitol and PHB show a high profit margin, achieving that the net profit margin in both scenarios be positive. Although, due to the increment in depreciation, utilities and operating costs the profit margin for the second scenario is 97% lower. For the base case, the electricity unit cost is 0.634 USD/kW, which is higher than the commercial price of this utility (0.11 USD/kW).

Table 4.

Figure 3.

3.4.Environmental Assessment

The environmental assessment is based on the criteria of the potential environmental impacts, described above. The results of the potential environmental impact per ton of products are presented in Fig. 4. These Figures shown that the best scenario, from the environmental point of view, is the one that considers cogeneration of the solid residues. According to Fig. 2, human toxicity by ingestion (HTPI) and terrestrial toxicity (TTP) potentials affect the most to the environmental impacts. The HTPI and TTP potentials are high due to organic waste present in liquid streams leaving the process.

Figure 4.

4. Conclusions

According to the results, the olive stone is an interesting material to be used as feedstock under a biorefinery concept to obtain value-added products. Despite of furfural and ethanol plants are not feasible by itself, part of the cost of these plants are covered by the other ones, thus the total feasibility is positive with a profit margin of 53%, for the second scenario. On the other hand, a cogeneration considered in scenario 3 is not feasible inside the biorefinery because of the additional capital cost. Regarding to the environmental analysis, the results reveals that scenario 2, which includes a cogeneration system, has the lower contamination level.

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Figure 1

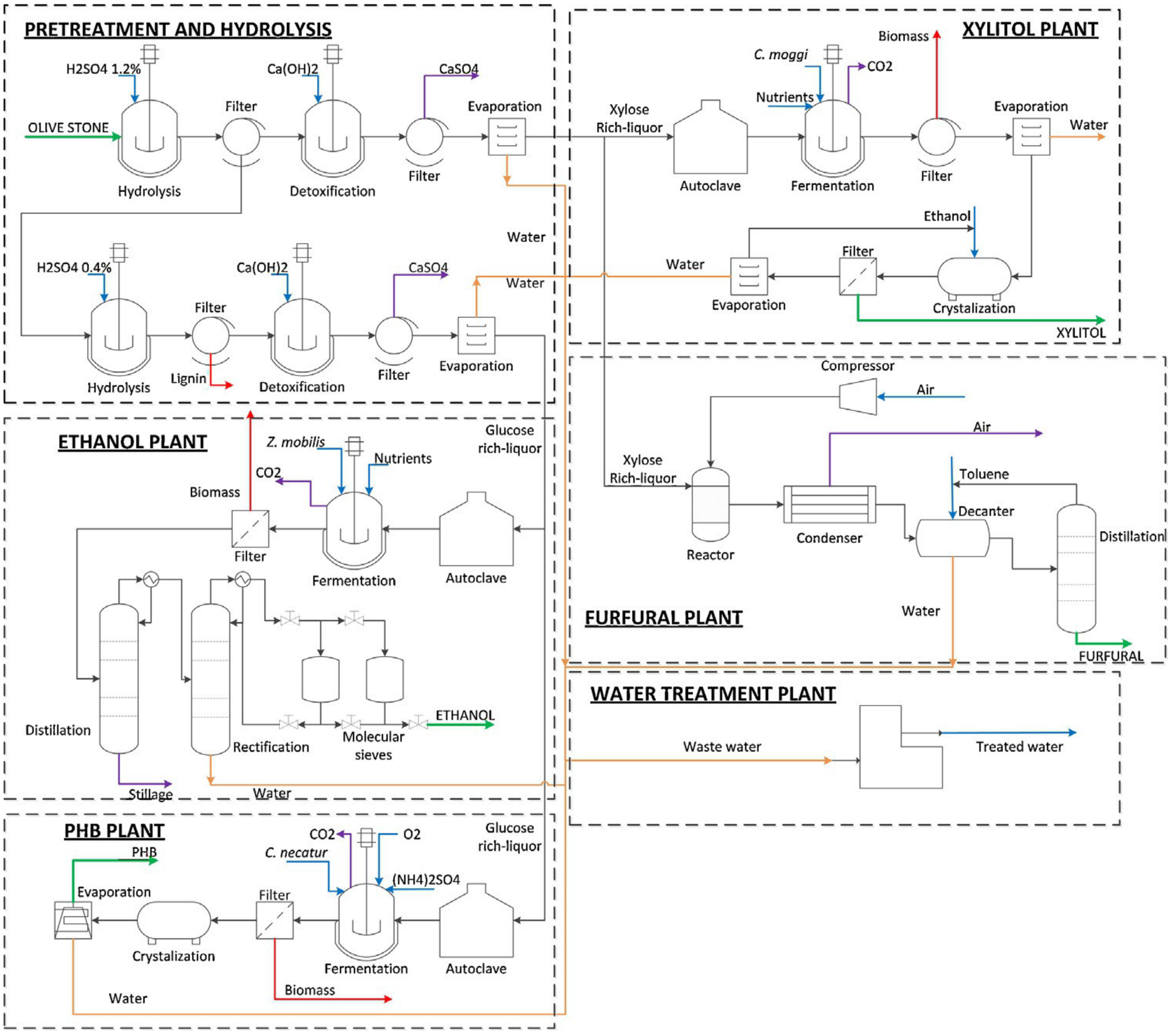

Fig. 1. Process flowsheet for scenario 1.

Figure 2

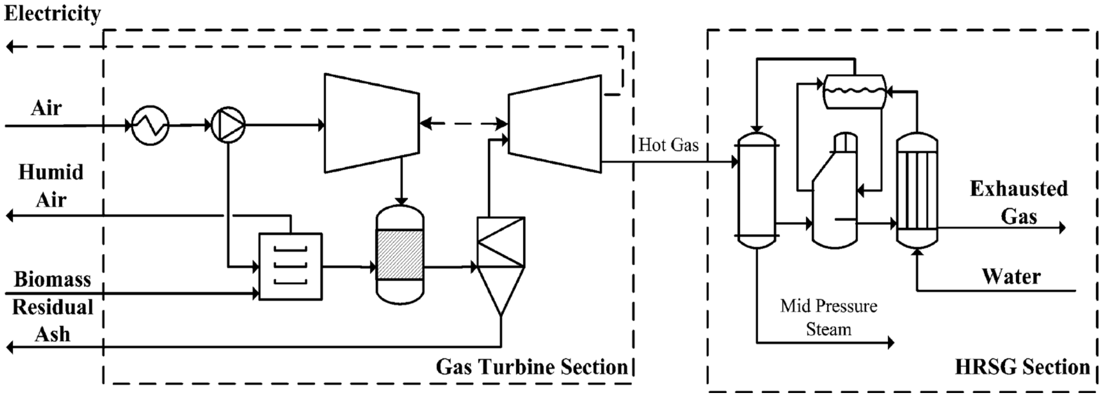

Fig. 2. Cogeneration process integrated to the biorefinery scheme in the second scenario.

Figure 3

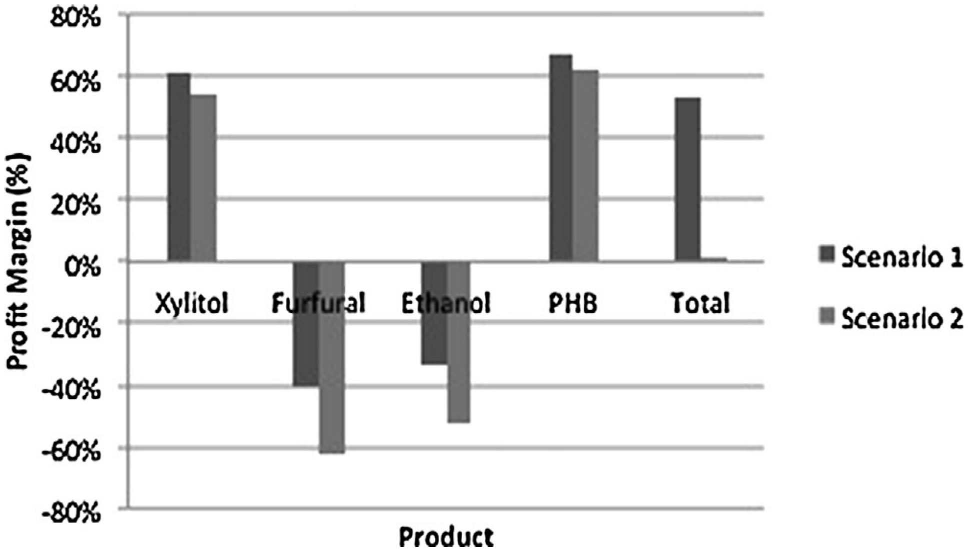

Fig. 3. Unit and net profit margin.

Figure 4

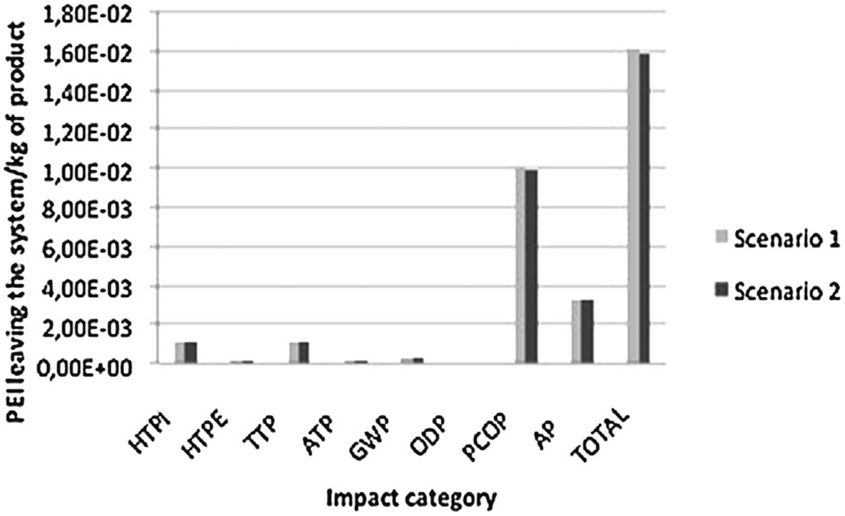

Fig. 4. Potential environmental impact per kg of product.

Table 1

Table 1 Olive stone composition used for simulation procedure.

Compound	%
Cellulose	20.10
Hemicellulose	29.92
Lignin	38.87
Extractives	10.54
Ash	0.57
Total	100

Moisture content: 7.71%.

Table 2

Table 2 Production capacities.

Product	Productivity
Xilitol	12.00 ton/day
Furfural	8.04 ton/day
Ethanol	18.20 m^3 /day
PHB	4.07 ton/day
Electricity	218 MW[a] 36 MW[b]

[a] Base Case

[b] Scenario 2

Table 3

Table 3 Annualized costs for each scenario.

Item	Base case		Scenario 1		Scenario 2	
	Million US$/year	Share (%)	Million US$/year	Share (%)	Million US$/year	Share (%)
Depreciation of capital	1.765	4	2.166	4	2.815	4
Raw material	–	–	11.491	19	11.491	16
Utilities	20.670	45	14.789	24	18.818	27
Operating	23.867	52	31.916	53	36.756	53
Total	46.302		60.363		69.880	

Table 4

Table 4 Unit production costs.

Product	Production cost				
	Base case	Scenario 1		Scenario 2	
Xilitol	–	3.12	USD/kg	3.61	USD/kg
Furfural	–	2.82	USD/kg	3.25	USD/kg
Ethanol	–	1660.00	USD/m^3	1890.00	USD/m^3
PHB	–	2.31	USD/kg	2.66	USD/kg
Electricity	634 USD/MW	–		–	