

LIGNOCELLULOSIC MULTI-PRODUCT BIOREFINERY: EVALUATION OF ECONOMIC AND ENVIRONMENTAL HOTSPOTS AND VALUE CREATION

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ABSTRACT: The slow progress in the commercialization of lignocellulosic biorefineries involving the biochemical conversion pathway points at the immaturity of existing biorefinery concepts. The current research suggests a systematic analysis of value creation chains and their economic and environmental hotspots within a multi-product biorefinery, having as the primary goal to promote sustainable biorefining processes. A lignocellulosic biorefinery producing ethanol, crude lignin oil and electricity was analyzed. Our methodology involves the transformation of a technological biorefinery model into the input-output one and the use of matrix notation to track the accumulation of economic and environmental attributes along biorefinery value chains. Three categories of indicators are chosen to reveal the most critical processing stages of every value chain, namely: (i) monetary and environmental profiles, (ii) the role of feedstock and (iii) infrastructure required to support main processes. Principal biorefinery hotspots refer to the biomass pretreatment, solids recovery and lignin drying stages, which is due mainly to wastewater treatment accompanying these processes. The other hotspot, lignin solvolysis technology, shows the environmental advantage of decreasing methanol content in the produced lignin oil. The proposed method is worth being applied to the analysis of new biorefinery concepts and new integration pathways.

Keywords: Multi-product biorefinery, hotspots, value chain analysis, life cycle assessment, techno-economic analysis

1 INTRODUCTION

The development of sustainable biorefinery plays a key role in the successful transition to bioeconomy. By definition, biorefining refers to “the sustainable processing of biomass into a spectrum of biobased products and bioenergy” [1]. Yet, the slow progress in the commercialization of lignocellulosic biorefineries involving the biochemical conversion pathway points at the immaturity of existing biorefinery concepts and pushes towards the analysis aimed at the identification and improvement of the most critical biorefinery processes, or hotspots.

Given further concerns about sustainable biomass conversion routes, more studies become available regarding biorefinery hotspot analysis. While the life cycle assessment (LCA) methodology involving standardized procedures to account for the environmental impact of products is widely used for the environmental assessment of biorefineries, techno-economic analysis (TEA), that converts technology-related data into the cost-related one, is applied for the evaluation of economic hotspots [2]. However, LCA of biorefineries is often disconnected from TEA [3]. Here, the term “hotspot” can be determined as a point where “major environmental problems or costs” appear on a product's life cycle [4].

By definition, a biorefinery involves a series of processing steps aimed at adding value to each intermediate output. Quite often, the main conversion processes are to be complemented with a comprehensive infrastructure serving all energy and ecological needs of a biorefinery due to its typically remote location. As a result, the value creation process may boost costs and environmental burdens added at each processing step, and this requires a uniform assessment of related hotspots.

In the current study, we suggest a systematic approach for the evaluation of hotspots of a stand-alone multi-product biorefinery, and involve the matrix

notation for this analysis. Thus, while most of studies focus on biorefinery hotspots with final products in sight, the proposed methodology allows for the value-specific analysis of said hotspots, including the evaluation of the role of infrastructure in those value chains.

The proposed approach is aimed mainly to complement the already existing TEA and LCA of a multi-product biorefinery, as, given its uniformity, this can be applied to a variety of indicators capable of revealing most critical processing stages. A lignocellulosic biorefinery simultaneously producing ethanol, crude lignin oil and electricity is selected in this study for demonstration purposes.

2 METHODOLOGY

Principal steps of the methodology applied in the current study are presented in Fig. 1.

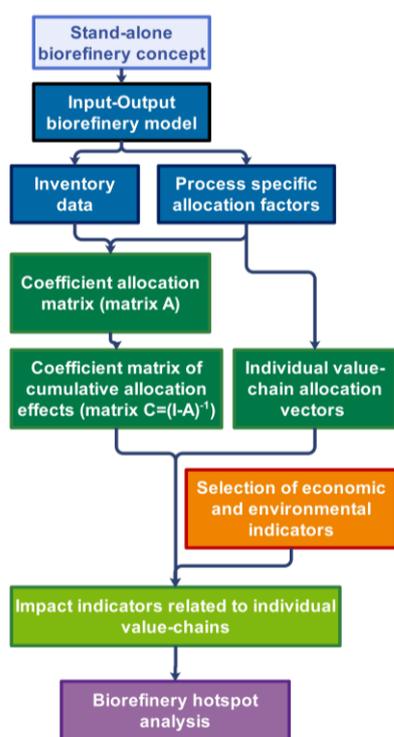


Figure 1: Methodological framework

2.1 Stand-alone biorefinery concept

We have considered a lignocellulosic biorefinery that simultaneously produces three co-products, namely: ethanol, crude lignin oil (CLO) and electricity.

The reference biorefinery model co-producing ethanol and electricity was developed by the *National Renewable Energy Laboratory, NREL* [5, 6], and the lignin-to-CLO valorization technology has been integrated into that design, using the Aspen Plus® software. Principal parameters of the CLO technology were provided by Vertoro, a Dutch company [7].

The biorefinery under consideration aims at processing about 104.2 ton of corn stover per hour with the ethanol production capacity of 22276 kg/h [5]. The conversion process includes biomass pretreatment technology, where corn stover is subjected to high pressure steam and dilute sulfuric acid that allows to convert hemicellulose carbohydrates into soluble sugars, as well as to reduce cellulose chain length and crystallinity [5]. After pretreatment, slurry undergoes enzymatic hydrolysis with produced on-site cellulase enzymes, and fermentation [5].

On leaving the fermentation area, beer is, first, subjected to distillation, where solid lignin fraction separated at the column bottom, while ethanol obtained at the top of the latter is further de-watered and purified.

In order to serve biorefinery energy needs, about 62% of the moist lignin residue is sent to combined heat and power (CHP) plant, and the remaining lignin stream is, first, dried to up to 0.8% (w.b.) and, then, subjected to mild solvolysis under the temperature of 200°C, pressure of 38 bar and using methanol as a solvolytic medium [7]. The solubilization process allows for the separation of the lower molecular weight lignin [7], which is referred to in this paper as CLO, while the insoluble higher molecular weight fraction (char) is sent back to the CHP plant for energy recovery. After this, CLO contains 77.5 wt% of

lignin, 10.0 wt% of methanol and about 1.6 wt% of water, and features a lower heating value of 24.2 MJ/kg that makes it suitable for the biofuel market, as well as for other value added applications. CLO hourly production capacity amounts to 3499 kg.

Along with the main conversion processes, the biorefinery features on-site production of energy (steam and electricity) and comprises all other infrastructure necessary for running biorefining processes.

Fig. 2 demonstrates the input-output representation of the biorefinery.

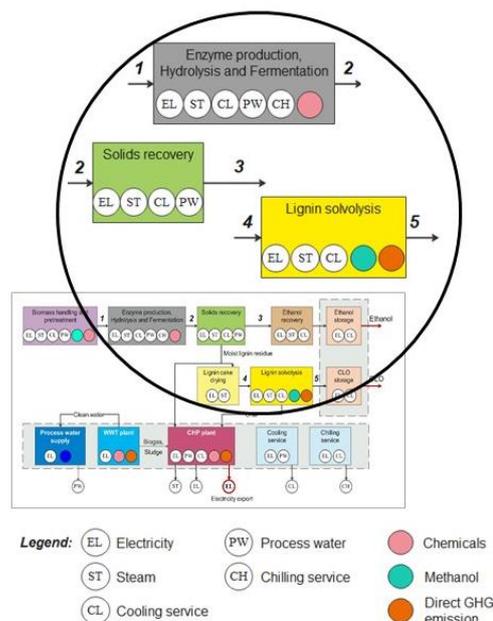


Figure 2: Representation of biorefinery model in input-output form

2.2 Matrix approach

We apply the matrix approach allowing to track the transfer of any process specific attributes along biorefinery value-chains, where the attributes can belong to either economic or environmental group of parameters (e.g. cost or emission).

The methodology includes several steps, namely: (i) the accounting of cumulative attributes belonging to intermediate streams and final products, (ii) the assignment of cumulative attributes to product-specific value-chains, and (iii) the analysis of hotspots of the said value-chains.

2.2.1 Cumulative attributes

A biorefinery represents a system of interconnected processes, and, thus, the first methodological step should be to transform the technological biorefinery model into the input-output one. However, the level of details would be defined by data availability and by the study goal.

If a process results in multiple outputs, the appropriate allocation method is to be used to assign attributes to each output. Clearly, the sum of process-related allocation factors is equal to one. In the end, a non-negative square coefficient allocation matrix can be developed for such system.

The development of the coefficient allocation matrix (matrix *A*) for the biorefinery and its properties were described in detail in [8]. The matrix maps the transfer of

attributes from flows to processes and vice versa for the entire biorefinery.

It was shown that the power expansion equation can be used to track inputs and outputs along the supply chain, as it is capable to capture both direct and indirect inputs [9]:

$$\mathbf{x} = \mathbf{y} + \mathbf{A} \cdot \mathbf{y} + \mathbf{A}^2 \cdot \mathbf{y} + \mathbf{A}^3 \cdot \mathbf{y} + \dots = (\mathbf{I} - \mathbf{A})^{-1} \cdot \mathbf{y} \quad (1)$$

where \mathbf{A} is $n \times n$ input-output matrix, \mathbf{x} is $n \times 1$ vector of output level, \mathbf{y} is $n \times 1$ vector of final deliveries, \mathbf{I} is the $n \times n$ identity matrix.

For a biorefinery case, since the effect of the transfer of attributes from one flow to another is defined via the multiplication of transfer coefficients, the \mathbf{A}^2 matrix will show attributes allocated due to the secondary transfer effect and so on, that results in the next equation [8]:

$$\mathbf{e}_c = \mathbf{e} + \mathbf{A} \cdot \mathbf{e} + \mathbf{A}^2 \cdot \mathbf{e} + \mathbf{A}^3 \cdot \mathbf{e} + \dots = (\mathbf{I} - \mathbf{A})^{-1} \cdot \mathbf{e} = \mathbf{C} \cdot \mathbf{e} \quad (2)$$

where:

- $\mathbf{e}_c = (e_{c1}, e_{c2}, \dots, e_{cn}) \in \mathbb{R}^n$ is the vector representing cumulative attribute (economic or environmental) related to the streams within biorefinery boundary;
- $\mathbf{e} = (e_1, e_2, \dots, e_n) \in \mathbb{R}^n$ is the vector of input attribute (economic or environmental);
- \mathbf{A} is the non-negative coefficient allocation matrix;
- $\mathbf{C} = (\mathbf{I} - \mathbf{A})^{-1}$.

The matrix $(\mathbf{I} - \mathbf{A})^{-1}$ in eq. 1 and eq. 2 is the Leontief inverse coefficient matrix. Here, this matrix reflects the share of cumulative attributes to be assigned to each flow within the system boundary.

It should be noted also that there is a clear analogy observed between the Leontief inverse and inverse of the technology matrix in LCA [10].

2.2.2 Individual value chains

In order to select sustainable biomass valorization routes, we should also obtain figures showing the accumulation of attributes inside individual value chains of biorefinery products. The latter can be obtained via the multiplication of a diagonal allocation matrix $\text{diag}(\mathbf{y}_k)$ specific to such value chain by the vector of cumulative attribute:

$$\mathbf{v}_k = \text{diag}(\mathbf{y}_k) \cdot \mathbf{e}_c \quad (3)$$

where \mathbf{v}_k is the vector of a cumulative attribute allocated to the k^{th} product value-chain; \mathbf{y}_k is the k^{th} product value-chain allocation vector. In this vector, each element is a product of the downstream non-zero allocation factors related to specific value-chain. The element of this vector related to the final product will be equal to one, and elements, which do not belong to an value chain equal zero.

2.2.3 Analysis of economic and environmental hotspots

We suggest several economic and environmental attributes that can be uniformly involved in the systematic analysis of biorefinery hotspots. These attributes are aimed at (i) the evaluation of monetary and environmental profiles of value chains, (ii) the analysis of feedstock role, and (iii) the analysis of the role of infrastructure required to support the main processes in

those value chains. These attributes are explained in more detail below.

(i) Production cost and GHG emission through individual value chain

Production cost and environmental impacts associated with a biorefinery product can be considered as the main metrics of its sustainability. This concept can be applied to all production stages through the value chain of each individual biorefinery product:

The production cost after each biorefinery process (PC_{ik}) is defined as:

$$\text{PC}_{ik} = (\text{E}_i + \text{OPEX}_i) / \text{P}_k \quad (4)$$

where PC_{ik} is normalized production cost after the i^{th} biorefinery process; E_i is the equivalent annual cost of the i^{th} process; OPEX_i is the operational expenses related to the i^{th} process; and P_k is the amount of final product related to the k^{th} value chain.

Equivalent annual cost E can be found using the next equation:

$$\text{E} = \text{DR} \times (1 + \text{DR})^n / ((1 + \text{DR})^n - 1) \times \text{TCI} \quad (5)$$

where DR is the discount rate; n is the project lifetime and TCI is the total capital investment.

Concerning environmental impacts, in this study we evaluate only greenhouse gas (GHG) emission, as it is the most widely adopted environmental metric of a biorefinery [11 - 13], however, any other impact categories can be analysed if needed.

The relative increase of an attribute at any process stage (RI , %) is defined as:

$$\text{RI} = (\text{Attribute increase at a process stage} / \text{Attribute increase in a value chain}) \times 100 \quad (6)$$

(ii) Accumulation of impact related to biomass in the individual value chain

Within the biorefinery, biomass is the primary source of bio-molecules to be converted to value-added products, but also it is the source of energy required by main conversion processes.

Ultimately, because energy is supplied from biomass-sourced residuals, the accumulation of biomass allocated along the value creation chains only expands, imposing also an inevitable increase in cost and boosting environmental impact associated with feedstock in these value chains.

The relative increase of biomass accumulation at any process stage compared to the total increase in a value chain is found using eq. 6.

(iii) The cost and GHG emission associated with the main and infrastructural processes

In our study, we separate the cost and the impact associated with the main and infrastructural processes and we attempted to analyse which particular main processes are more responsible for the use of infrastructure. For that, we refer to the concept allowing to define a portion of equipment as settled inside battery limits (ISBL), or outside battery limits (OSBL), and we analyse the ratio of OSBL to ISBL for installed cost of equipment, operational expenses and GHG emission associated, and apply these indicators to the value creation chain analysis.

According to [14], the factor range of OSBL for a stand-alone new greenfield plant may account for up to 60 – 120% of process costs. However, since this indicator has not been reported for the environmental analysis, the same range is adopted for the analysis of GHG emission hotspots in the current study.

2.3 Tools and methods

We used the SimaPro® (v.8.0.2) software to access the background process-related life-cycle inventory databases. The IPCC 2013 v.1.03 impact assessment method with a 100-year time horizon was used for the quantification of GHG emission. The R software [15] was used to convert the coefficient allocation matrix into the coefficient matrix of cumulative allocation effects. The Aspen Plus® software was used to integrate the lignin conversion technology into the reference model and to access technology-related data.

3 RESULTS

Fig. 3 and 4 show the frequency of appearance of main biorefinery processes in the set of the most critical ones, secondary critical ones, and so on, and summarize the results of this categorisation for three value chains.

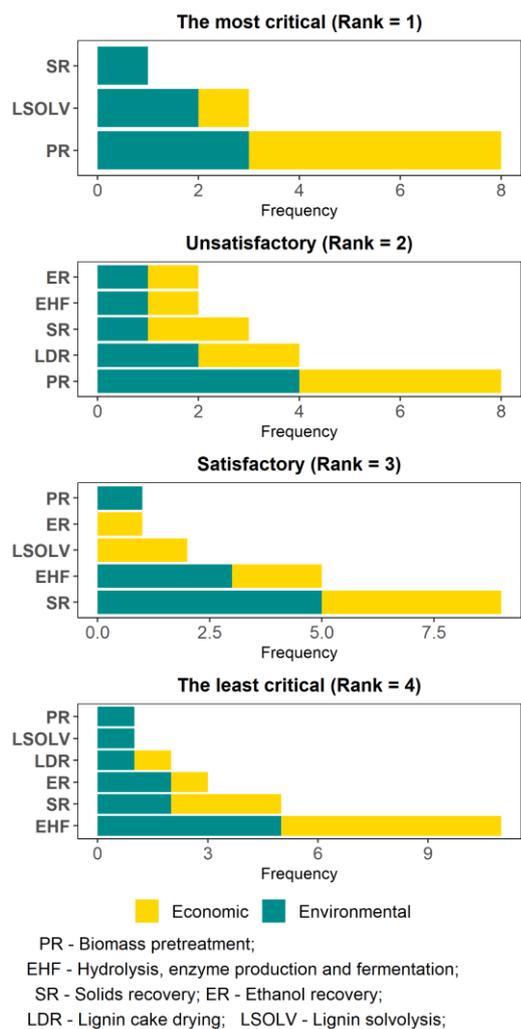


Figure 3: Ranking analysis

Value chain	Relative increase, %			OSBL / ISBL, %		
	A	B	C	D	E	F
Ethanol value chain						
Pretreatment hydrolysate	39%	45%	36%	149%	43%	137%
Beer	30%	26%	11%	85%	27%	82%
Diluted ethanol	22%	23%	44%	104%	34%	105%
Ethanol	9%	6%	10%	102%	35%	111%
Lignin-based value chain						
Pretreatment hydrolysate	19%	18%	16%	149%	43%	137%
Beer	15%	10%	5%	85%	27%	82%
Moist lignin residue	11%	9%	20%	104%	34%	105%
Dry lignin residue	13%	9%	22%	123%	34%	122%
Crude lignin oil	41%	53%	37%	110%	29%	56%
Electricity value chain						
Pretreatment hydrolysate	16%	17%	26%	149%	43%	137%
Beer	13%	10%	8%	85%	27%	82%
Moist lignin residue	9%	9%	31%	104%	34%	105%
Electricity	61%	64%	35%	433%	128%	294%

A - Production cost
 B - GHG emission
 C - Accumulation of impact related to biomass
 D - OSBL/ISBL Installed equipment cost ratio
 E - OSBL/ISBL Operational expenses ratio
 F - OSBL/ISBL GHG emission ratio

R=1 The most critical process
 R=2 Unsatisfactory process
 R=3 Satisfactory process
 R=4 The least critical process

Figure 4: Impact of processes in individual value chains

As it can be seen from Fig. 3, three processes persist among the most critical, with the pretreatment technology leading this category. Although the remarkable role of the pretreatment technology in the total capital investment of a cellulosic biorefinery was highlighted in [16], the current analysis also uncovers the significant impact of this technology on production cost and GHG emission of the ethanol value chain and the crucial role of infrastructure required to support the pretreatment process (Fig. 4). Wastewater treatment (WWT) facility dominates in both cost and emission associated with services required for the biomass pretreatment step, that also supports the observation concerning a considerable contribution of the digester and supplement treatment to the total capital costs of a lignocellulosic biorefinery [17]. The results obtained demonstrate that environmental and economic performance of the biorefinery under review will benefit mainly from an improvement in the pretreatment technology as this will positively affect the most critical stages in all three value chains.

The second process that emerges very frequently on the list of the most critical biorefinery processes is lignin solvolysis. Although the GHG emission profile of CLO obtained allows to decrease GHG emission by 76% compared to fossil fuel and, hence, to comply with the threshold set by the European Renewable Energy Directive [18], this profile can benefit further from the reduction of highly GHG intensive methanol content in CLO. However, another hotspot can be expected because of existing trade-off between virgin solvent and thermal energy needed for the solvent recycling [19].

Solid recovery, including beer distillation and lignin filtration processes, can be regarded as another hotspot of the biorefinery (Fig. 3), as it showed the worst performance in terms of accumulation of impact related to biomass in the ethanol value-chain (Fig. 4), and it was found unsatisfactory in terms of accumulation of impact related to biomass and the cost of infrastructure involved.

The drying of lignin residue can be also regarded as a biorefinery hotspot (Fig. 3), as, although it occurred in the second critical rank category, it showed unsatisfactory results in the accumulation of impact related to biomass, as well as in all indicators related to the economic and environmental performance of

infrastructure (Fig. 4). As it follows from results, the installed cost of CHP plant amounts to 56% of the total services involved, and both WWT and CHP facilities play notable role in the GHG emissions associated with drying step.

We can also mention enzymatic hydrolysis, along with enzymes production, and fermentation, as well as ethanol recovery as unsatisfactory in both economic and environmental sense. While hydrolysis influenced mainly production cost and GHG emission of the ethanol value chain (Fig. 4), the ethanol recovery bunch of processes was the second in terms of operational expenses and GHG emission associated with infrastructure, where WWT facility took the leading role in the environmental impact associated with recovery process.

Several recommendations can be elicited based on the hotspot analysis for the considered biorefinery.

Given the crucial role of infrastructure required to support biomass pretreatment process, the biorefinery concept would significantly benefit from a decrease in the load of solids at the pretreatment step. As the main role of the pretreatment technology is to decompose the rigid structure of biomass, liberating lignin and making hemicellulose and cellulose available for the downstream hydrolysis, the removal of lignin prior to the hydrolysis step would decrease the amount of water needed for the downstream valorisation of the named carbohydrates. The lignin-based value chain can also benefit from this conceptual change, as the lignin cake drying step, the other biorefinery hotspot, will be eliminated from the biorefinery model. A decrease in the amount of water would also positively affect the solid recovery step (also identified as a hotspot), as less energy will be required for distillation and evaporation processes.

4 LIMITATIONS

The suggested approach imposes several limitations that need to be taken into consideration along with hotspot analysis. The main one refers to the allocation procedure. For instance, the recent study highlighted the importance of specific allocation methods applied to CHP plant [8]. It should be noted that allocation procedure was regarded to as one of the main sources of uncertainty in the attributional LCA [20].

Another limitation is related to the selection of thresholds for the analysis of economic and environmental indicators. For instance, while there is understanding of a reasonable range of installed equipment cost referred to OSBL of a stand-alone new greenfield plant, there is no such data available for rating environmental impacts or operational expenses of infrastructural facilities.

5 CONCLUSIONS

The current study suggests a systematic approach aimed at the analysis of value creation chains and their economic and environmental hotspots within a multi-product biorefinery.

The methodology involved the transformation of a technological biorefinery model into the input-output one and the use of matrix notation to track the accumulation of economic and environmental attributes along the biorefinery value creation chains. Thus, while most of

studies focus on biorefinery hotspots with final products in sight, the proposed methodology allowed for the process-specific evaluation of economic and environmental hotspots. Three categories of indicators have been chosen to reveal the most critical processing stages, namely indicators allowing for: (i) the evaluation of monetary and environmental profiles of product value chains, (ii) the analysis of the role of feedstock, and (iii) the analysis of the role of the main processes and infrastructure in those value chains.

Our study highlights a conspicuous role of infrastructure required to support the biomass pretreatment, solids recovery and lignin drying processes, especially when it comes to wastewater treatment in the biorefinery, and proposes improvements of the biorefinery concept based on earlier separation of lignin-containing stream.

Moreover, although the GHG emission profile of lignin oil obtained allows to decrease GHG emission by 76% compared to fossil fuel, it is concluded that a significant environmental improvement can be attained in the lignin-based value chain via a reduction of methanol content in the lignin oil; however, “energy versus recycled solvent” trade-offs must be analysed in line with the analysis of economic and environmental profiles of that value chain.

The study results demonstrate the effectiveness of the matrix approach for the analysis of value creation and of hotspots within biorefinery. The proposed method is worth being applied to the analysis of new biorefinery concepts and new integration pathways. Among limiting factors, we stress the expected sensitivity of results to allocation procedures that should be properly addressed during analysis.

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