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Wood Chips Components Separation with a New Wet-Milling Process Compared to Chemical Depolymerization: A Technical, Economic, and Environmental Comparison

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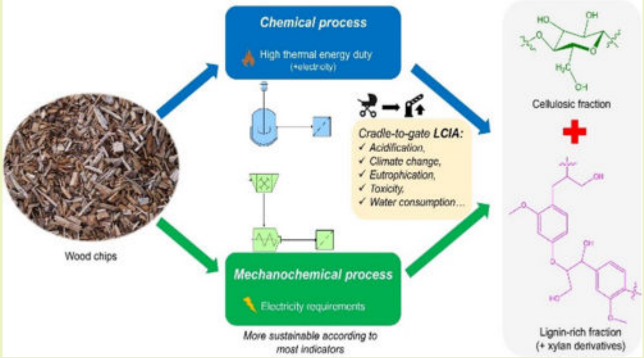
ABSTRACT: This article evaluates two processes, wet milling and chemical depolymerization, for the end-of-life of wood waste in terms of environmental performance (ex ante life cycle assessment), energy balance, and economic analysis of the operating costs. Cellulose, hemicellulose, and lignin are essential components with numerous applications. The study provides valuable insights for industry stakeholders, policymakers, and researchers of the wet milling process (WMP), which is scarcely reported in the literature. The chemical depolymerization process (CDP) is discontinuous and more energy-intensive, while the WMP is a continuous reaction demanding milder conditions and shorter times. However, the milling process requires a pretreatment to reduce the wood chip size. Economic analysis shows that the CDP has lower operational costs when considering the average European electricity price in 2019. This is a result of the price differences between steam and electricity. For lower electricity prices such as in France or using utility-scale solar photovoltaics, the WMP has lower operational costs. The WMP also outperforms the CDP in most environmental indicators, such as global warming potential, particularly when using green electricity technologies.



KEYWORDS: *cellulose, economic analysis, life cycle assessment, wet milling, lignin, wood chips*

INTRODUCTION

Wood fibers are complex, hierarchically structured natural materials that are mainly constituted by cellulose, hemicellulose, and lignin.[1,2] As each constituent has a chemical function, they are not homogeneously distributed in lignocellulosic fibers. Concretely, the outer cell wall layers, particularly the middle lamella, are mainly composed of lignin (60−70 wt %) and hemicellulose (20 wt %), while the cellulose content ranges from 10 to 20 wt %, on average.[3] The inner layers, namely the primary and the secondary walls, are much richer in hemicellulose and cellulose.[3] Amorphous constituents at the outer layers of fibers enable the binding between the different fibers in the lignocellulosic biomass, which leads to this hierarchically organized structure.[4] Today, several methods are used to decompose this naturally assembled structure, encompassing mechanical, thermomechanical, chemical, and combined approaches.[5] The choice of separation technique depends on the purpose.

For instance, cellulosic pulps are currently used in several sectors, papermaking being the most demanding, and their production usually implies the removal of lignin in several stages, such as cooking, fibrillation, and/or bleaching.[6,7] On a smaller scale, cellulose is used for higher value-added products; such is the case of cellophane or rayon, or additives for different sectors, namely, cosmetics, paints, biomedicine, and the food industry.[8−10] Cellulose is also used in the textile sector, and besides the traditional usage of cotton, recent research keeps unveiling its potential for sustainable and functional fabrics.[11] Moreover, the potential of cellulose and hemicellulose for producing second- and third-generation biofuels (particularly bioethanol) should be highlighted.[12] The market for lignin-based products is still smaller than that of cellulose, but they have been proven effective for fertilizing, sand-fixing, as bioactive compounds, and as adhesives, among other possibilities.[13−15]

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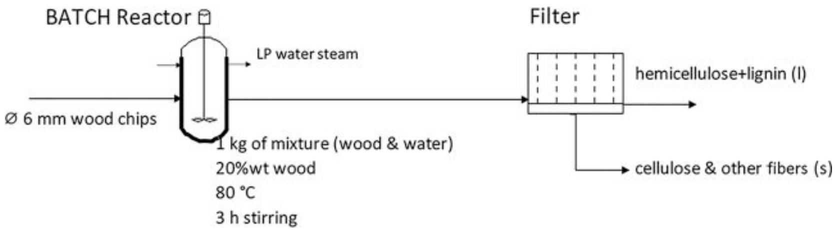

Figure 1. Flowchart of the chemical depolymerization for fiber extraction.[15]

The need for increasing the sustainability of industrial operations for papermaking, lignin extraction, or any other purpose implies processing wood fibers, which initiated the development of greener processes, where the reduction of energy and decrease in consumption of chemicals is pursued. Wet milling is a process of chemical reactions induced by mechanochemical action that can usually be operated at room or moderate temperatures. These processes attracted attention because of their advantages compared with traditional processes such as the chemical depolymerization process (CDP) in terms of energy savings, process intensification, and higher productivity.[16−18] However, there is limited understanding of the mechanochemical processes because they are usually conducted in closed devices, which are often regarded as black boxes.[19] Polymer mechanochemistry has been a topic of great interest over the last years, dealing with organic solvent reactions and operating with dissolved polymers.[20] On the other hand, the solid mechanochemistry of polymeric materials lacks generality, mainly because, in the case of thermoplastic polymers, they absorb most of the mechanical energy for melting. In this sense, the mechanochemistry of wood-based biopolymers, such as the case of cellulose and hemicellulose/lignin, is of great industrial relevance.[16]

The main advantage of applying mechanochemical or wet milling processes (WMPs) to lignocellulosic materials is the increase of the available surface of fibers (due to size reduction effects) while providing the action of chemical reagents.[21] This operation can not only be conducted in several devices, such as ball mills (planetary or vibrator), attritors, grinders, or high-pressure and ultrasonic homogenizers but also in continuous flow millers and extruders.[16] Each piece of equipment will impart specific forces over the lignocellulosic biomass, leading to different process efficiency and product characteristics.

In the present work, we propose a novel method for wood waste treatment and fiber separation. The WMP is evaluated by means of an ex ante life cycle assessment (LCA) and economic analysis of the operating costs, and compared to an extraction process, the CDP that was previously reported by Shuai et al.[15]

Ex ante LCAs are used to inform decision-making during the early stages of product development or technology innovation, help stakeholders understand potential environmental consequences, and identify opportunities for improvement. In an ex ante LCA, researchers estimate the environmental impacts based on available data, assumptions, and models related to the future implementation of the product or process under study. As it deals with future scenarios, results are estimative, and uncertainties may be higher compared to retrospective LCAs, which assess existing products or processes. Nevertheless, ex ante LCAs can provide valuable insights for decision-makers, enabling them to make informed choices about the development and adoption of new technologies or processes with a focus on sustainability.[22,23]

Even though some articles regarding the use of mechanochemistry or wet milling as an example of avoiding other environmentally intensive processes have been recently published,[24−27] the scientific literature is scarce of LCAs of these processes. A comprehensive literature review of research articles performing LCA of fiber separation methods of wood chips by WMP or CDP was conducted using a rigorous search methodology by strings:

- (lca OR "life cycle assess*" OR "life cycle analy*" OR lci OR "Life cycle*") AND (cellulose OR lignin OR hemicellulose) AND (mechanochem* OR mechano-chem*)
- (lca OR "life cycle assess*" OR "life cycle analy*" OR lci OR "Life cycle*") AND (cellulose OR lignin OR hemicellulose) AND "chemical depolymeri*"

The search was conducted in Web of Science and Scopus, one of the most extensive scientific databases. This search yielded only 9 results for the first string and 13 for the second string. Filtering for only research articles resulted in no findings for both strings. This emphasizes the novelty and significance of the present paper.

Process Description. *Chemical Depolymerization Process.* The extraction stage of the process reported by Shuai et al.[15] operates in a batch reactor at a liquid:solid ratio of 4:1, using 6 mm diameter wood chips as a source of biomass. This operation takes place at 80 °C for 3 h under agitation, in an acidic medium, and in the presence of formaldehyde. At an industrial scale, the heating of the reactor is typically provided with low-pressure (LP) water steam, according to the operating conditions of the process. The last step consists of cake filtration that separates the solid from the liquid fraction. According to the authors, the liquid fraction mainly contains lignin and derivatives of xylan (often, the main constituent of hemicellulose), while the solid fraction is mainly composed of glucans (attributed to cellulose) and a small fraction of lignin.[15] Figure 1 provides a flow diagram of the CDP used as a base scenario for the present study.

Wet Milling Process. The WMP consists of a high-efficiency bead mill filled with ZrO_2 (stabilized with 20% CeO) micromilling beads, DYNO-MILL MULTI LAB, which is dedicated to continuous flow wet-milling application. It uses the collision of zirconia microbeads with the reactants as the main principle to activate the reaction. This mechanical energy only requires indirect use of electricity, which is supplied to the motor to motion the rotor and discs of the machine. The microbeads housed in the milling chamber represent, by volume relative to the total volume of the stationary chamber, from 55 to 70%. They are substantially spherical, with a mean diameter ranging from 0.5 to 1 mm and a Vickers hardness measured according to standard EN ISO 6507-1 typically ranging from 1000 to 1400 HV1.[28]

This process is still in its design phase, and the preliminary tests are performed at a laboratory scale provided the most optimal conditions presented in the paper. The WMP works at liquid:solid ratios between 50:1 and 10:1, operating at temperatures ranging from 25 to 80 °C for 5 to 60 min.

The consumption of reagents for a certain yield and the filtration step are considered equivalent to the CDP, even though it is believed that the lower concentration of wood might allow for an easier separation depending on the filtration technology and the properties of the cake.[29]

The WMP (Figure 2), in the laboratory, requires a pretreatment with a hammer mill to reduce the size of the wood chips from 6 to 1 mm in diameter. The size reduction before the DYNO-MILL MULTI LAB helps to avoid accumulation and therefore gives a narrower residence time which can be assimilated to the liquid flow rate presented.

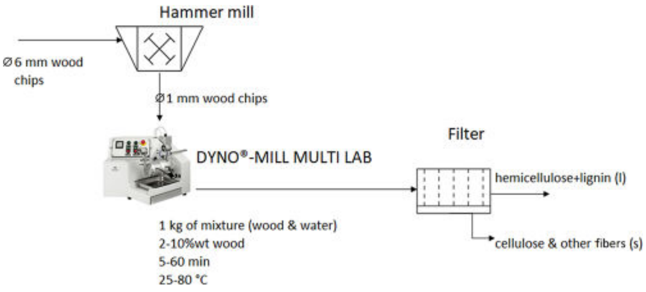

Figure 2. Flowchart of the WMP for fiber extraction.

When scaling up the process to pilot or industrial scale, using a bigger mechanochemical reactor, possibly makes this pretreatment no longer necessary. However, as this hypothesis has not yet been fully validated and remains with a conservative approach, the energy demand in the hammer mill step is considered in the following sections. In addition, specific surface changes could also have an impact on the process efficiency.

The main issue to be addressed is whether the energy input of the pregrinding step and the lower wood concentration inside the reactor are outweighed by the advantages of the continuous WMP. These advantages include savings achieved by reducing the operating temperature and the residence time.

METHODOLOGY

Life Cycle Assessment. LCA is a widely used systematic tool that estimates the environmental impacts of products or production processes considering every stage in their life cycle.[30−33] According to the framework presented in the ISO 14040 (2006) and ISO 14044 (2006), an LCA is regulated in four phases: goal and scope, life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation.

The product sustainability solutions software Sphera LCA for experts (version 10.7.1.28) is being used to quantify the environmental impacts of WMP and CDP by making use of its extensive databases. The product environmental footprint (PEF) has been selected to perform a multicriteria measurement of the environmental performance of the processes. This method has been developed by the European Commission (2018) as one of the building blocks of the flagship initiative of the Europe 2020 Strategy—"A Resource-Efficient Europe".[37] The midpoint indicators were weighted following the same methodology from the European Commission to evaluate their relevance for the study and which categories are more important in the evaluated system. Weighting in the LCA is a step in the interpretation phase that involves assigning relative importance or significance to different impact categories based on value choices or preferences. It helps to consolidate and simplify the results of an LCA, making it easier to compare alternatives and providing informed decision-making.[38,39]

The functional unit of the study is used as the basis of the calculation. In this article, the scope of the study is treating wood waste by breaking down wood chips into cellulose, hemicellulose, and lignin. Therefore, the functional unit is the quantity of wood waste introduced into the reactor. The fact that we are comparing a batch process (CDP) with a continuous process (WMP) makes selecting a consistent functional unit a challenge. Taking the direct CDP as a basis, its flow rate has been calculated at 0.0667 kg/h (0.2 kg of wood fed to a 3 h batch). In the case of the WMP, the technology operates with a different wood fraction and residence time than the CDP; in other words, with different flow rates. To ensure that the study was conducted under the same baseline, with the same wood input and output for both the WMP and CDP, an allocation factor was used. The allocation factor was based on the ratio between the flow rate of the CDP and the flow rate of each scenario of the WMP. This method of defining the flow rate has been discussed and implemented in various publications.[21,40−44] Another example of a similar approach is the consideration of module D within the EN 15804:2012 + A2:2019 for the inclusion of information beyond the cradle-to-gate scope in construction product LCAs.[45,46] These allocation factors are presented in Table 1 for 11 different scenarios defined from the operating ranges presented in Wet milling processsection.

Table 1. Allocation Factors Were Applied to the WMP Scenarios

process	scenario	wood fraction flow rate, kg/h	allocation factor[a]
CDP	average values	0.067	1.0
WMP	(1) average values	0.10	0.67
WMP	(2) minimum wood fraction	0.040	1.7
WMP	(3) maximum wood fraction	0.20	0.33
WMP	(4) minimum reaction time	0.50	0.13
WMP	(5) maximum reaction time	0.050	1.3
WMP	(6) highest temperature	0.10	0.67
WMP	(7) room temperature	0.10	0.67
WMP	(8) maximum milling energy consumption	0.10	0.67
WMP	(9) minimum milling energy consumption	0.10	0.67
WMP	(10) 0.77 mm final particle size	0.24	0.28
WMP	(11) 3 mm final particle size	0.040	1.7

[a] Allocation factor = CDP flow rate/WMP flow rate (for each scenario).

The study's stages included in the LCA correspond to the production of the raw material, the thermal energy needed to heat the reaction mixture, and the mechanical energy needed to move, mix, mill, or stir the wood waste. This is a cradle-to-gate study. The boundaries of both processes are represented in Figure 3.

The LCI is then utilized to gather data on raw materials, energy, and emissions and determine environmental impacts. We have presented the material flows and operating conditions for each scenario in Table 2, while Table 3 displays the energy flows.

The results are projected with the present electricity mix in Europe (Table 9), the projected electricity mix in 2030 (Table

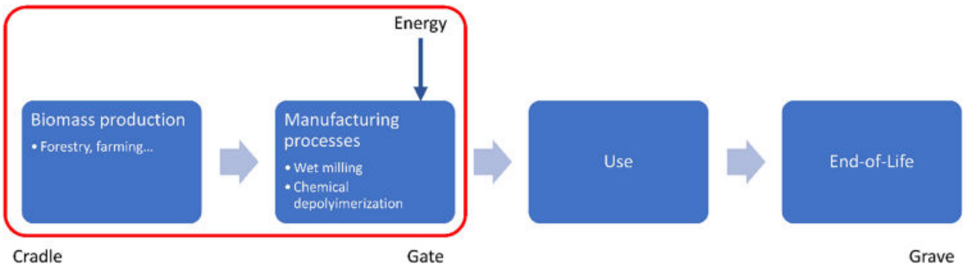

Figure 3. Boundaries of the LCA.

Table 2. Material Flows and Operating Conditions (1 kg Mixture Inside the Reactor)[a]

process	scenario	water fraction (wt %)	wood fraction(wt %)	Cp_{mix} (J/g °C)	time (h)	T_{max} (°C)
CDP	average values	80	20	3.7	3.0	80
WMP	(1) average values	95	5.0	4.1	0.50	52
WMP	(2) minimum wood fraction	98	2.0	4.2	0.50	52
WMP	(3) maximum wood fraction	90	10.	4.0	0.50	52
WMP	(4) minimum reaction time	95	5.0	4.1	0.10	52
WMP	(5) maximum reaction time	95	5.0	4.1	1.0	52
WMP	(6) highest temperature	95	5.0	4.1	0.50	80
WMP	(7) room temperature	95	5.0	4.1	0.50	25
WMP	(8) maximum milling energy consumption	95	5.0	4.1	0.50	52
WMP	(9) minimum milling energy consumption	95	5.0	4.1	0.50	52
WMP	(10) 0.77 mm final particle size	88	12	3.9	0.50	52
WMP	(11) 3 mm final particle size	98	20	4.2	0.50	52

[a] Scenarios 10 and 11 have been tested with higher wood fraction, which presented difficulties in operating the reactor and worse quality in the final product. Therefore, as presented in Figure 2, best operating conditions have been defined as a wood fraction between 2 and 10 wt %.

Table 3. Energy Balance of the Different Scenarios under Study

process	scenario	energy used heating (kJ)	energy used stirring/milling (kJ)	energy used in pretreatment (kJ)	energy consumption (kJ)
CDP	average values	290	1.76		292
WMP	(1) average values	99.0	24.0	52.5	184
WMP	(2) minimum wood fraction	250	60.0	52.5	372
WMP	(3) maximum wood fraction	48.0	12.0	52.5	121
WMP	(4) minimum reaction time	20.0	4.80	52.5	85.2
WMP	(5) maximum reaction time	200	48.0	52.5	308
WMP	(6) highest temperature	202	24.0	52.5	287
WMP	(7) room temperature		24.0	52.5	84.8
WMP	(8) maximum milling energy consumption	99.0	38.4	52.5	198
WMP	(9) minimum milling energy consumption	99.0	19.2	52.5	177
WMP	(10) 0.77 mm final particle size	41.0	8.00	78.5	128
WMP	(11) 3 mm final particle size	247	60.0	14.5	322

10), and the projected electricity mix in 2050 (Table 11). These projections are derived from the "EU Reference Scenario 2016—Energy, Transport and GHG Emissions—Trends to 2050" published by the European Commission (2016).[47] The environmental factors used for the different electricity mix are available in the Sphera professional databases.[48]

Material. 6-mm diameter lodgepole pine wood chips were previously dried in an oven at 50 °C to a moisture content of 11.7 wt % and stored in hermetic plastic bags at room temperature. This is the raw material used by Naimi et al.[49] Naimi et al.[49] reported the energy consumption equations that are used in this study to estimate the energy balance of the pretreatment step of the WMP. For the sake of comparison, similar wood chips are assumed as raw materials in both WMP and CDP.

Pregrinding Setup. The WMP needs to reduce the size of the wood chips from 6 to 1 mm in diameter to be able to process the wood in the DYNO-MILL MULTI LAB. According to the material and required size, this may be performed using a hammer mill equipped with swing hammers and powered by a three-phase induction motor at a speed of 3490 rpm.[49,50]

Naimi et al.[49] designed a series of experiments for different screen sizes with different initial and final particle sizes.[49] Energy consumption during this operation was measured and reported as summarized in Table 4. For the present work, solely the values for the initial size of wood chips, 6 mm in diameter, and their final particle size, 1 mm, have been utilized. Specific energy was linearly interpolated from the values reported in Table 4 according to eq 1.

Table 4. Specific Energy of the Grinding Pretreatment for the WMP[a]

ID	d_{fp}[b] (mm)	d_{op}[c] (mm)	specific energy (kW h/t)
1	9.7	1.1	16
2	9.7	0.77	25
3	3.6	1.1	8.2
4	3.6	0.77	19

[a] Data extracted from the paper published by Naimi et al.[49] [b] d_{fp}: diameter of the feed particles. [c] d_{op}: diameter of the output particles.

$$\text{Linear Interpolation }(y) = y_1 + \frac{(x - x_1) \times (y_2 - y_1)}{(x_2 - x_1)} \tag{1}$$

where x_1 and y_1 are the first coordinates; x_2 and y_2 are the second coordinates; x is the point to perform the interpolation; y is the interpolated value.

The linear interpolation formula is the simplest method that is used for estimating the value of a function between any two known values. We calculated y = specific energy by interpolating rows 1 and 2 of Table 4, for x = diameter of the output particles = 1 mm. Then, we repeated the interpolation with rows 3 and 4 of Table 4. Finally, we interpolated both results for x = diameter of the fed particles = 6 mm.

Based on eq 1 and the interpolated data from Table 4, we assume 14.57 kW h/t (52,454 J/kg) as the average energy consumption of the pretreatment in the hammer mill.

Energy Balance. The energy consumption of the rest of the equipment was estimated with the advanced process calculations published by Piccinno et al.[51] The energy input in the CDP is therefore the sum of the thermodynamic equations for stirring and heating energy, which includes the energy needed to reach the reaction temperature and the energy needed to compensate for the heat loss. The energy input for the WMP is the energy associated with the specifications of a hydro-micromilling reactor, which is 8−16 kW h/t. The energy input is consistent with the range provided by Piccinno et al.[51] for a ball miller. As mentioned in Grinding Set-Up, the filtration step has been considered equivalent for both processes, and thus, it is excluded from the comparative analysis.

Economic Analysis of the Operating Costs. Regarding the economic comparison, raw material costs for treating 0.0667 kg/h of wood waste are the same for both processes, and therefore, they have been excluded from the comparative analysis. Therefore, the point is to evaluate if the pretreatment will reduce the particle size before the mechanochemical reactor is viable despite increasing the operational costs. This might be the case because of the expected savings in the reaction step compared to those of the CDP.

Most recent data at the time of performing the study was used in the analysis. Low-pressure steam price of 4.44€/100 kW h according to Turton et al.[52] without credit for power has been assumed for the supply of heating energy in both processes. To compare the importance of electricity prices on process costs and the variability of current energy markets, three different scenarios have been considered in a sensitivity analysis. The electricity prices which have been used are (i) 21.59€/100 kW h, the average electricity price in Europe in 2019 according to Eurostat, (2021);[53] (ii) 17.65€/100 kW h, the average electricity price in France in 2019,[54] and (iii) 5.98€/100 kW h, corresponding to the electricity costs from utility-scale solar photovoltaics (PV) in 2019, according to the International Renewable Energy Agency (IRENA) report from the year 2020.[55]

■ RESULTS AND DISCUSSION

Energy Balance. The overall energy demand, along with the energy demand for heating, stirring, or milling, and the pretreatment (if any) of the CDP and the different scenarios of the WMPs are presented in Table 3.

Therefore, CDP is more energy-intensive in 8 of the 11 scenarios studied. This is due to the high energy demand for heating the mixture to 80 °C and keeping this temperature for 3 h. According to the operating conditions (80 °C), this heat is typically supplied with LP water steam (on an industrial scale). On the other hand, the WMP has a higher electricity demand for grinding and milling the wood. The main challenge of the WMP relies on the optimization of the wood fraction introduced in the reactor, as inferred from the results of scenario 2. The key to this process is to take advantage of its lower operating temperature and residence time, as we can learn from the results of scenarios 4 and 7. Finally, results from scenarios 10 and 11 indicate that it is advisable to spend more energy reducing size in the pretreatment than in the reaction itself. The energy requirements for the pretreatment step of the WMP have been plotted in Figure 4 to evaluate its tendency. As can be seen, the increase in the energy demand is exponentially correlated to the reduction of the particle size.

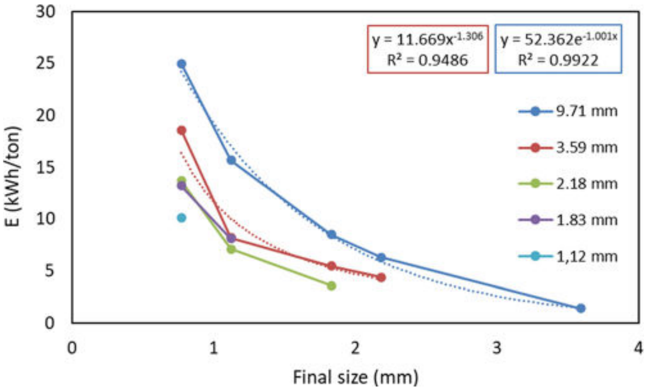

Figure 4. Energy demand depending on final particle size.[49]

Economic Analysis. The energy consumption is used to determine the operational cost of the processes. This was calculated for three different alternatives. Table 5 presents the operational costs for a European average electricity price in 2019, Table 6 presents the operational costs for a French average electricity price in 2019, and Table 7 uses the electricity costs from utility-scale solar PV.

The CDP requires thermal energy, besides reagents such as formaldehyde and HCl, whose consumption is assumed not to impart a difference in the scope of the study. However, it can be relevant to study energy usage associated with distillation to purify the final product if required.[56] This is expected to be supplied with LP water steam, which is significantly more inexpensive than the electricity required by the WMP. Assuming as electricity price the average electricity price in Europe in 2019 (Table 5), even though the CDP is more energy-intensive, its operational costs are lower than those of the WMP. When considering the electricity price in France in 2019 (Table 6), which is 18% lower, we already observe one scenario (4: minimum reaction time) that is favorable for the WMP. Finally,

Table 5. Economic Balance for European Average Electricity Price, 2019

process	scenario	European electricity price (€/3000 h)	total cost (€/3000 h)
CDP	average values	0.320	11.0
WMP	(1) average values	15.3	18.9
WMP	(2) minimum wood fraction	21.7	30.9
WMP	(3) maximum wood fraction	13.1	14.9
WMP	(4) minimum reaction time	11.8	12.5
WMP	(5) maximum reaction time	19.6	26.9
WMP	(6) highest temperature	15.3	22.7
WMP	(7) room temperature	15.3	15.3
WMP	(8) maximum milling energy consumption	17.8	21.5
WMP	(9) minimum milling energy consumption	14.4	18.0
WMP	(10) 0.77 mm final particle size	15.6	17.1
WMP	(2) minimum wood fraction	13.4	22.5

Table 6. Economic Balance for French Average Electricity Price, 2019

process	scenario	LP water steam cost (€/3000 h)	French electricity price (€/3000 h)	total cost (€/3000 h)
CDP	average values	10.7	0.260	10.9
WMP	(1) average values	3.62	12.5	16.1
WMP	(2) minimum wood fraction	9.22	17.8	27.0
WMP	(3) maximum wood fraction	1.76	10.7	12.5
WMP	(4) minimum reaction time	0.720	9.64	10.4
WMP	(5) maximum reaction time	7.32	16.0	23.3
WMP	(6) highest temperature	7.40	12.5	19.9
WMP	(7) room temperature		12.5	12.5
WMP	(8) maximum milling energy	3.62	14.6	18.2
WMP	(9) minimum milling energy	3.62	11.8	15.4
WMP	(10) 0.77 mm final particle size	1.51	12.7	14.3
WMP	(11) 3 mm final particle size	9.06	11.0	20.0

Table 7. Economic Balance for Utility-Scale Solar PV Average Electricity Price, 2019

process	scenario	LP water steam cost (€/3000 h)	Utility-scale solar PV electricity price (€/3000 h)	total cost (€/3000 h)
CDP	average values	10.6	0.09000	10.7
WMP	(1) average values	3.62	4.23	7.85
WMP	(2) minimum wood fraction	9.22	6.02	15.2
WMP	(3) maximum wood fraction	1.76	3.63	5.39
WMP	(4) minimum reaction time	0.720	3.27	3.99
WMP	(5) maximum reaction time	7.32	5.42	12.7
WMP	(6) highest temperature	7.40	4.23	11.6
WMP	(7) room temperature		4.23	4.23
WMP	(8) maximum milling energy	3.62	4.94	8.57
WMP	(9) minimum milling energy	3.62	3.99	7.61
WMP	(10) 0.77 mm final particle size	1.51	4.31	5.82
WMP	(11) 3 mm final particle size	9.06	3.72	12.8

regarding the electricity costs from utility-scale solar PV, 7 of the 11 scenarios are already favorable for the WMP. Those are 1: average values, 3: maximum wood fraction, 4: minimum reaction time, 7: room temperature reaction, 8: maximum milling energy consumption, 9: minimum milling energy consumption, and 10:0.77 mm final particle size.

It is important to consider the direction in which the electricity mix is moving in the different countries of the European Union[47,57] and the increasingly strict restrictions on the use of fossil fuels and polluting processes. This could make the WMP even more interesting in the coming years.

Moreover, after comparing the deviation of the operational cost for each scenario, we can determine which variables have a greater effect on the final cost to prioritize them in the optimization stage. Table 8 presents the variability of the operational cost, depending on the configuration of different operating conditions. The maximum, minimum, and average value of the result in each operating condition has been categorized as worst, average, or best, depending on the operational costs generated in each case. Those operating conditions with higher variability are those that should be prioritized as it means that they have a greater influence on operating costs. In addition, the "best" column indicates the costs associated with the best economic scenario for each variable, with the minimum reaction time (scenario 4) being the most inexpensive process setup.

Table 8. Variability of Operational Costs Depending on Each Factor

operating conditions	worse (€/3000 h)	best (€/3000 h)	variability (%)
wood fraction	30.9	14.9	85%
reaction time	26.9	12.5	76%
temperature	22.6	15.3	39%
particle size	22.5	17.1	29%
milling energy	21.5	18.0	18%

Environmental Analysis. Tables 9−11 gather the environmental midpoint impacts according to the PEF methodology. The calculation has been made for the 11 scenarios using the results of the CDP as a basis for a percentage. Those impacts higher than 100% mean that they are categories in which the CDP has better environmental performance (color-coded in red). On the other hand, those midpoint indicators with less than 100% indicate that they are categories in which the WMP is environmentally better (color-coded in green). In the case of an environmental impact equal in both processes, this has been identified in yellow.

Table 9. LCIA of the WMP over the Impacts of the Chemical Process (Present Time)

Scenarios	1 (%)	2 (%)	3 (%)	4 (%)	5 (%)	6 (%)	7 (%)	8 (%)	9 (%)	10 (%)	11 (%)
Acidification	44	44	62	38	53	48	41	47	44	73	35
Climate Change	57	92	53	36	84	77	38	61	56	60	77
Ecotoxicity, freshwater	82	90	95	71	95	83	81	90	79	111	65
Eutrophication, freshwater	125	140	123	117	136	126	125	131	123	130	120
Eutrophication, marine	38	37	56	33	46	43	34	40	38	66	31
Eutrophication, terrestrial	38	36	55	32	45	42	33	39	37	65	30
Human toxicity, cancer	57	75	63	42	75	68	46	61	55	72	61
Human toxicity, non-cancer	60	79	64	45	78	72	48	63	58	72	66
Ionizing radiation	196	252	194	158	242	196	195	223	186	229	163
Land Use	27	13	52	27	28	27	27	28	27	62	12
Ozone depletion	190	243	188	154	234	190	189	216	181	221	160
Particulate matter	30	19	53	28	32	31	29	31	30	63	17
Photochemical ozone formation	37	35	55	31	44	42	33	39	36	64	30
Resource use, fossils	61	99	55	38	90	82	40	65	59	61	83
Resource use, mineral & metals	120	166	116	91	156	132	108	134	115	134	119
Water use	120	125	114	119	121	120	120	121	120	113	123

Considering that most of the indicators are colored green, this anticipates that the WMP is environmentally better in most cases. Both processes demand similar amounts of raw material. So on, the advantage of WMP is mainly due to the use of electricity instead of steam.

Most of the midpoint indicators of the WMP get better results with the implementation of green electricity technologies. This is due to the higher use of electricity compared to the CDP. Nevertheless, some indicators such as water use, use of minerals (zirconia), and ozone depletion remain better in the CDP.

To evaluate the significance of the midpoint indicators that perform worse in the WMP, these indicators have been weighted using the European Commission's Environmental Footprint (EF) 3.0 methodology.[36] Among these indicators, water use, mineral use, and ozone depletion have relatively low weights, while land use and fossil resource use are more significant. This is probably due to the intensive use of energy for heating or milling the mixture and due to the use of wood as a raw material. In this case, while the carbon emissions of the wood chips are primarily biogenic, land use takes a significant role. The

Table 10. LCIA of the WMP over the Impacts of the Chemical Process (2030)

Scenarios	1 (%)	2 (%)	3 (%)	4 (%)	5 (%)	6 (%)	7 (%)	8 (%)	9 (%)	10 (%)	11 (%)
Acidification	42	41	60	36	50	46	38	45	41	70	33
Climate Change	52	84	49	32	77	72	32	55	51	55	73
Ecotoxicity, freshwater	75	80	89	65	86	76	74	81	72	103	59
Eutrophication, freshwater	140	161	136	128	156	141	140	149	137	144	133
Eutrophication, marine	36	34	54	31	43	41	32	38	36	64	29
Eutrophication, terrestrial	36	33	53	30	42	40	31	37	35	63	28
Human toxicity, cancer	61	80	66	45	80	72	50	65	59	76	65
Human toxicity, non-cancer	58	77	63	44	76	71	47	62	57	71	65
Ionizing radiation	117	140	127	97	141	117	116	131	112	149	94
Land Use	29	15	53	28	30	29	29	29	28	64	13
Ozone depletion	226	296	219	183	280	226	226	258	215	258	192
Particulate matter	30	19	53	28	32	31	29	30	30	63	17
Photochemical ozone formation	35	32	53	30	42	40	31	36	35	62	28
Resource use, fossils	56	93	51	34	84	78	35	60	55	57	80
Resource use, mineral & metals	144	201	137	110	187	156	133	163	138	159	140
Water use	120	124	114	119	121	120	120	120	119	112	123

calculated weights for all the midpoint indicators can be found in Table 12.

CONCLUSIONS

An ex ante LCA has validated that the WMP exhibits superior environmental performance for most of the studied midpoint indicators compared to the CDP. Furthermore, its prospects appear more promising, largely due to its reliance on mechanical energy rather than thermal energy. However, some midpoint indicators demonstrate better results for the CDP process, although these indicators are less important in the weighting assessment. By maximizing the wood fraction introduced into the reactor and recycling the water used in the process, even better results for the water use indicator can be achieved.

Regarding the economic analysis and, considering the possibility that, in a scaling up of the process, the requirements of the pretreatment with the hammer mill would not be so demanding, it can be anticipated that the results obtained correspond to a lower limit of the energy costs of the WMP, which still needs further validation at industrial scale.

Table 11. LCIA of the WMP over the Impacts of the Chemical Process (2050)

Scenarios	1 (%)	2 (%)	3 (%)	4 (%)	5 (%)	6 (%)	7 (%)	8 (%)	9 (%)	10 (%)	11 (%)
Acidification	40	38	58	34	47	44	36	42	39	68	31
Climate Change	46	75	44	27	69	66	26	48	45	48	67
Ecotoxicity, freshwater	70	73	85	62	80	71	69	75	68	99	55
Eutrophication, freshwater	151	177	144	136	168	151	150	161	147	155	143
Eutrophication, marine	35	32	53	30	41	39	31	36	35	62	28
Eutrophication, terrestrial	34	31	52	29	40	39	30	35	34	61	27
Human toxicity, cancer	65	87	70	49	86	77	54	71	63	81	69
Human toxicity, non-cancer	57	76	62	44	75	69	46	61	56	70	64
Ionizing radiation	104	122	116	88	125	105	104	116	100	136	83
Land Use	29	16	54	28	31	29	29	30	29	64	14
Ozone depletion	332	447	309	264	417	332	332	383	315	365	285
Particulate matter	29	18	52	28	31	30	28	30	29	62	16
Photochemical ozone formation	34	30	52	29	40	38	29	35	33	61	27
Resource use, fossils	51	86	47	31	78	73	31	55	50	52	75
Resource use, mineral & metals	196	274	181	150	253	208	185	224	187	212	185
Water use	119	124	113	118	120	119	119	120	119	112	122

Consequently, the WMP is a promising innovative alternative that, depending on the electricity price, may be performed at lower operational costs than direct chemical extraction.

As presented in the article, shifting to mechanochemical systems can provide numerous benefits, such as improving efficiency and selectivity, reducing the need for solvents and thermal energy, or enhancing scalability. The fact that the WMP requires electricity and the CDP requires thermal energy highlights the environmental potential of the WMP. With the greenification of electricity, as forecasted in the European Green Deal, this assessment might increase.

However, further experimentation is required to determine how promising the mechanochemical approach is compared to an acidic formaldehyde-assisted CDP with no mechanical fractionation. In future research, we aim to optimize the scenarios that demonstrate superior performance in terms of energy consumption, economic evaluation, and LCA. In addition, by scaling these scenarios up to pilot or industrial

Table 12. Midpoint Indicators Weight

midpoint indicator	weights[a] CDP	WMP
acidification	0.0030%	0.0040%
climate change	3.1%	4.1%
ecotoxicity, freshwater	1.3%	2.2%
eutrophication, freshwater	ca. 0%	ca. 0%
eutrophication, marine	0.0010%	0.0010%
eutrophication, terrestrial	0.011%	0.010%
human toxicity, cancer	ca. 0%	ca. 0%
human toxicity, noncancer	ca. 0%	ca. 0%
ionizing radiation	0.058%	0.15%
land use	75%	61%
ozone depletion	ca. 0%	ca. 0%
particulate matter	ca. 0%	ca. 0%
photochemical ozone formation	0.0030%	0.0030%
resource use, fossils	20.%	29%
resource use, mineral and metals	ca. 0%	ca. 0%
water use	1.1%	3.9%

[a] It should be emphasized that the weights assigned to the midpoint indicators are independent within each process. These weights serve to evaluate the relative importance of the different indicators within each process, rather than comparing the weight of a single indicator between the two processes.[34,35]

levels, we can obtain more realistic data, thereby enabling more comprehensive assessments and insights.

■ ASSOCIATED CONTENT

Data Availability Statement

The data sets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Author Contributions

We, the authors, hereby give our consent for the publication of this manuscript in the journal. S.A.: methodology, formal analysis, investigation, writing—original draft, and visualization. I.M.: conceptualization, validation, resources, supervision (new technology), and funding acquisition. A.B.: methodology, validation, writing—review and editing (environmental assessment). V.L.: conceptualization, investigation, and resources. R.X.: writing—review and editing (economic assessment). R.A.: writing—review and editing and validation. M.D.A.: writing—review and editing and supervision. J.P.: writing—review and editing. I.S.: writing—review and editing and validation. P.F.: methodology, validation, writing—review and editing, project administration, and funding acquisition.

Notes

We confirm that this is an original work and has not been previously published in any other scientific journal, nor is it under consideration for publication elsewhere. The authors have consulted the Guide for Authors before this submission. The manuscript has been prepared in compliance with the Ethics in Publishing Policy and with the rest of indications described in the aforementioned guide.
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