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Lignin as a renewable resource for bio-oil and oligomers production. An environmental perspective

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ABSTRACT

The depletion of fossil resources and climate change is already a fact that entails a great concern among the population. Efforts to search for renewable resource alternatives with low environmental impact is now a priority for authorities, industries and society in general. Added to this, there is the problem of the high demand for goods and services by the population, which continues to increase, leading to a high production of waste. In this context, the lignocellulosic biomass has a key role, as it is used in large industrial activities, such as the food industry, paper production and livestock farming, among others. To this end, the amount of lignin produced is very significant, which turns into the need to search for process alternatives for its recovery and valorization. Its calorific capacity allows a two-fold benefit, firstly because of its capacity to be used as a renewable fuel and for the production of natural chemicals, such as phenolic compounds, antioxidants, and bioplastics, and secondly because of the reduction in the amount of lignocellulosic waste released to the environment. This is the focus of this manuscript, where a large-scale modeling of a lignin valorization process to obtain bio-oil and oligomers is developed. In addition, to evaluate its suitability, an environmental study has been carried out, applying the Life Cycle Assessment methodology, and a techno-economic analysis as well. The results obtained have shown that the use of residual lignin as a raw material to obtain value-added products is viable both environmentally and techno-economically.

1. Introduction

The extended use of fossil-based fuels and products entails an environmental problem and an important concern about the future needs and demands (Wang et al., 2021). Based on the fact that the fossil resources are not infinite, there is an urgent need on looking for alternative feedstocks for the production of all the demanded products from the society, encompassing from the plastics, that are part

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Acronyms

CC	Climate Change
OD	Ozone Depletion
HT,c	Human Toxicity, cancer
AD	Acidification
ET	Eutrophication, terrestrial
EF	Eutrophication, freshwater
EM	Eutrophication, marine
TF	Ecotoxicity, freshwater
RM	Resource use, minerals and metals
RF	Resource use, fossils
FCI	Fixed Capital Investment
COM	Cost Of Manufacture
MSP	Minimum Selling Price

of most of the daily routine products and services (Basmage and Hashmi, 2020; Pouzada, 2021), to the fuels, which are the motor for the transportation of all the good and services required by the consumers (Hanif et al., 2022; Piggot and Erickson, 2022; Yildiz, 2018), and also the mean of transport for humans. Governments and laws are putting huge efforts to reduce the impact of the society daily lives on the environment, as climate change is now a reality that is affecting over the fauna, flora, water resources, air quality and human health (Bhatt, 2021; Letcher, 2021; Tong et al., 2022).

In this context, the use of lignocellulosic biomass has gained a huge attention. Its natural, aromatic macromolecule and renewable nature converts it in a sustainable alternative to fossil resources (Beckham et al., 2016; Karunarathna and Smith, 2020; Kim and Yoo, 2021). Even though huge amounts of lignin are produced in plenty of industrial facilities, almost 50 million tons of lignin are obtained just from paper and biorefinery facilities (Goliszek and Podkościelna, 2019; Li et al., 2021), as it is one of the essential components of biomass feedstocks, its molecular complexity reduce its use for the production and recovery of high-added value compounds, that is why most of the valorization pathways are based on energy recovery (Wang et al., 2019). This complexity is based on the aromatic units that forms part of the lignin molecular structure, linked by carbon-oxygen and carbon-carbon strong bonds which limit the accessibility and entailing a reduction on the biomass conversion and valorization. This is the reason why pre-treatment stages are required to enhance the availability of lignin as a renewable resource and convenient feedstock for alternative industrial process, looking for sustainability and environmental protection.

Although lignin is a generic term for this molecular component of biomass, it can certainly be classified according to the upstream process from which it is obtained. Considering that most of the lignin is obtained from the pulp production process, it can be classified according to the pulping process that has been carried out (Fernández-Rodríguez et al., 2019; Viikari et al., 2009). The process required for the separation of the cellulose available in the tree biomass, for the subsequent production of paper, requires a chemical pulping process, that, depending on its nature, it will be known as kraft, Organosolv or sulfite process, being these the most developed at industrial level (Bajpai, 2018, 2016; Young et al., 2003). The most widespread process for wood delignification is the kraft one, representing mostly an 80 % of the worldwide wood pulping procedure (Kun and Pukánszky, 2017; Peretti et al., 2015), leading to an annual production of more than 130 Mtons of kraft pulp (Bruijnincx et al., 2015). This method is based on the use of hydroxides and hydrosulfides chemicals, which are added to the wood pulp, requiring high temperatures (above 150 degrees Celsius) for breaking the glucoside bonds and allowing the separation of the cellulose (Reyes et al., 2016). The residual stream produced within this pulping process is well-known as black liquor, containing both the hemicellulose and the lignin (Bonhivers and Stuart, 2013; Vakkilainen, 2017). Looking for the separation of lignin, for its subsequent utilization as a renewable resource and feedstock for other industrial process, a precipitation with mineral acids could be developed, under a pH-controlled process (Mahmood et al., 2016). The most important advantage of this recovery procedure is that a very low among of residual sulfur is present on the separated product, about 1–2 %, entailing a reduced environmental load and an easily to handle lignin (Kun and Pukánszky, 2017).

Currently, most pulp industries consider black liquor as a source for energy production, which is considered a low-value valorization pathway. These recovery pathways range from biofuel production to the manufacture of bio-based chemicals, biopolymers, or, as proposed in this manuscript, bioadhesives and oligomers (Arias et al., 2024; Kumar and Verma, 2024; Morya et al., 2022). The benefits of combusting black liquor for energy production are related to its calorific potential. In some cases, the energy produced from lignin and hemicellulose in black liquor is sufficient to cover all or most of the energy needs of the pulp mill. This avoids the use of external fossil energy and can even generate a surplus of energy for sale to the electricity grid (IEA Bioenergy, 2007).

Another option for black liquor is its potential valorization into biochemicals. However, this requires an intensive pre-treatment to break down the lignin structures, which has a high energy demand and can pose environmental sustainability challenges (Hu et al., 2023; Sillero et al., 2021). Depolymerizing black liquor through pyrolysis, oxidation, or hydrogenolysis to obtain high-value phenolic compounds is also a possibility (Hernández Mañas et al., 2022; Zhou et al., 2022). However, improvements in these processes are necessary, as low selectivity and difficulties in phenolic separation reduce the economic viability and sustainability of this alternative. A similar challenge exists for producing aromatic compounds, such as vanillin or syringol, which requires chemical modification of lignin in black liquor—a process currently difficult to scale for commercial production due to low cost-effectiveness and limited

technological development (da Silva et al., 2009; Erdocia et al., 2012). In this context, catalytic-based processes that could reduce process time, improve economic viability, and avoid energy-intensive pre-treatment for lignin valorization may be a more viable alternative.

To this end, the recovery of the kraft lignin is the main focus of the present manuscript, looking for the production of bio-oil and oligomers by a base-catalyzed depolymerization procedure. Looking for establishing the environmental advantage of this procedure, together with the environmental loads that it entails and how the process could be improved, an environmental assessment is developed, considering the Life Cycle Assessment (LCA) methodology. On the other hand, even though sustainability and environmental awareness is important, also the economic viability has to be accounted for analyzing the suitability of the present process alternative for lignin recovery. In this sense, a techno-economic assessment (TEA) has also been considered, bearing in mind the market price fluctuation and the operational costs, among others.

The proposed pathway for producing bio-oil and oligomers through black liquor valorization aims to offer a sustainable and economically viable alternative to fossil-derived phenolic compounds, which are significant contributors to climate change due to greenhouse gas emissions. Replacing fossil-based phenolic compounds with bio-based, lignin-derived alternatives can help reduce their carbon footprint and support the achievement of net-zero emission targets, bioeconomy, and circular economy goals at the European level.

Current and upcoming regulations are increasingly encouraging the use of renewable materials and more circular production strategies, moving beyond energy valorization. This study aligns with these policies, offering an alternative that helps companies proactively meet emerging sustainability and circularity standards. From an economic viability, lignin used to produce phenolic compounds originates from a waste stream in the pulping sector, making it a renewable and relatively low-cost raw material. Technologically, the molecular structure of lignin-based products, such as bioadhesives and oligomers, is highly versatile and rich, increasing their applicability across various value chains, including construction materials, wood panels, and insulation products.

In this context, the findings of this manuscript could help value chain actors and stakeholders drive a paradigm shift in the pulp and paper production industries regarding black liquor valorization, while also encouraging industries reliant on fossil-derived phenol to consider transitioning to bio-derived phenolic compounds.

2. Methodology

2.1. Process description

The process of lignin recovery is based on a base-catalyzed depolymerization (BCD), in order to obtain the lignin fractions: oligomers and bio-oil. The procedure as proposed by Solt et al., (2018) has been used as a basis for performing the modeling, looking for developing the mass and energy balances required for developing the LCA methodology, and the selection of the equipment and stages needed, required for the TEA. Fig. 1 illustrate the global process of lignin recovery for bio-oil and oligomers production. The process starts by dissolving the lignin in a sodium hydroxide solution in mixing tank (V-101). The mixture following driven to continuous high-pressure reactor (FR-101) with 10 min residence time. The pressure increased into 250 bar thanks to hight pressure pump (HP-101), and the temperature was set to 320 °C. Under these conditions the hydrolysis takes places allows to obtain low molecular weight oligomers, due to the cleavage of the aryl-aryl and aryl-methyl ether bonds founded on the high molecular lignin structure. Afterwards the reaction process has ended, the temperature of the stream decrease by exchange energy with the inlet stream (E-101)

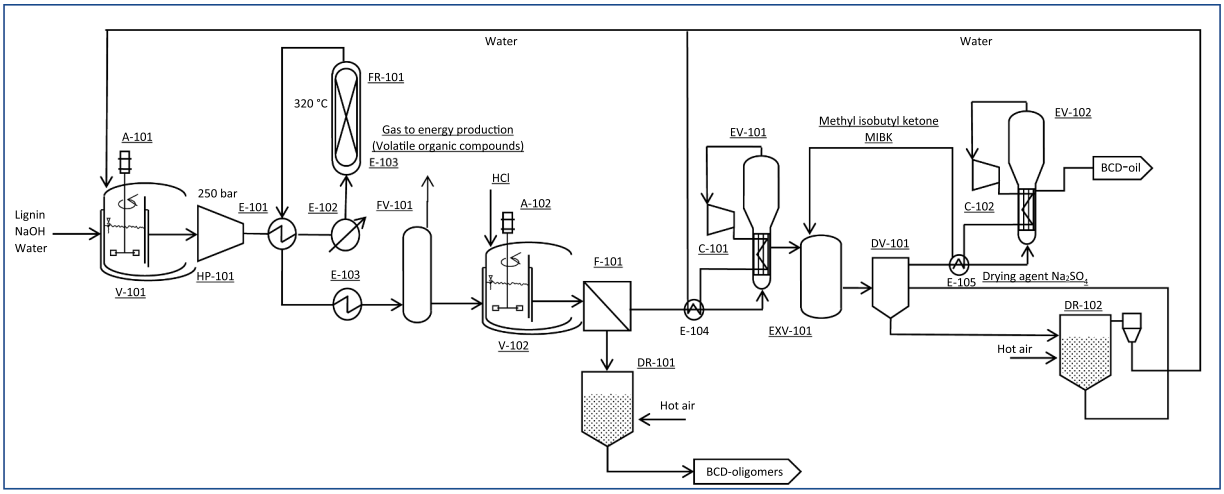


Fig. 1. Lignin recovery to produce bio-oil and oligomers: simplified process stages, main operation conditions and equipment required. Acronyms: HP (High Pressure Pump), E (Heat Exchanger), FR (Flow Reactor), DV (Decanter), F (Filter), DR (Dryer), V (Vessel), A (Agitator), FV (Flash Vessel), EXV (Liquide-Liquide Exchange Vessel).

and the pressure are decreased at ambient levels, to perform the downstream process. After the reaction process, except to main-stream also produced some volatile organic compounds (methanol, formic acid and acetic acid and carbon dioxide) where collected and send to co-generate heat and power station (CHP). This power station supply with electricity and steam the plant reducing its energy requirements. The CHP designed based on the Humbird et al. (2011). The downstream separation purification (DSP) stages are initialized with the precipitation of the BCD-oligomers under acidified conditions using hydrochloric acid and afterwards separated with a chamber filter press. Once the oligomers are separated, the rich in bio-oil permeate solution concentrated five times using a mechanical vapor recompress evaporator (MVR) (EV-101). The condensate water, from evaporation process, recirculated and the concentrated stream driven to liquid-liquid extraction (EXV-101) where the bio-oil separated from the aqueous solution using methyl isobutyl ketone (MIBK). After the liquid-liquid extraction, follows the decanting step (DV-101) where separated the organic from the aqueous phase. At this step, sodium sulfate salts used as a drying agent in order to reduce miscible of the MIBK into water. Then follows the bio-oil recovery step by evaporation of the MIBK in the MVR evaporator (EV-102) and recirculated to the liquid-liquid extraction unit. The salts dried using a spray drier (DR-102) and reused in the decanting step.

2.2. Preliminary techno-economic evaluation

The techno-economic evaluation was based on the experimental results of Solt et al., (2018). The mass and energy balances were developed using the process design software Unisim (Honeywell) considering annual production capacities of total products (BCD-oligomers and BCD-oil) between 10,000–100,000 t/year. The equations used for obtained the mass and energy balances required for developing of the life cycle inventory are as follows:

$$\text{Inputs} = \text{Outputs} \tag{1}$$

Eq.1 is considered for both mass and energy balances, in the case of the mass balances the unit of measurement is kg/h, while for the case of energy balances is kJ.

To calculate the energy requirements for each equipment input and output flows:

$$Q = m \cdot Cp \cdot \Delta T \tag{2}$$

In which Q is the heat energy (kJ), m is the mass flow (kg), Cp is the specific heat capacity of the flow (kJ/(kg·K) and ΔT is the temperature variation (K).

The sizing of each operation unit and each capacity was performed based on standard engineering procedures described in the literature (Peters et al., 2003; Ulrich and Vasudevan, 2004; Turton et al., 2018). The construction material of each process equipment is stainless steel and appropriate pressure factors were used on process steps where applied high pressure as from Turton et al. (2018). The free-on-board purchased equipment cost (Ceq.fob) estimated by taking into consideration the characteristic size of the units and the established textbooks and literature-cited reports (Turton et al., 2018., Dheskali et al., 2017; Humbird et al., 2011; Peters et al., 2003). The preliminary techno-economic methodology followed with accuracy up to ± 30 % has been described by Ioannidou et al. (2022a,b). The plant operation time was assumed on 7920h/y. The economic metrics Fixed Capital Investment (FCI) was estimated by multiplying the sum of the $C_{eq.fob}$ with a Lang factor of 5 (Dheskali et al., 2020). The Lang factor ranges from 3 to 6 when the $C_{eq.fob}$ is used for the estimation of the FCI (Peters et al., 2003; Turton et al., 2018). A Lang factor of 5 has been used in this study due to the construction of a new industrial plant using a high-risk new technology and the utilisation of expensive construction material in the process equipment. The cost of manufacture (COM) was estimated using the equation (Eq.3) proposed by Turton et al. (2018).

$$COM = 0.18 \times FCI + 2.73 \times COL + 1.23 \times (C_{UT} + C_{RM}) \tag{3}$$

where C_{OL} represents the operating labour cost, C_{UT} includes utilities expenses and CRM stands for the raw material expenses. The coefficients used in Eq. 1 have been estimated by Turton et al. (2018) in order to include the contribution of all secondary product manufacturing cost categories (e.g. maintenance, marketing, R&D) that can be associated to the main cost categories used in Eq. 1. The unitary cost of utilities, supplied by off-sites, are $9.45/t low pressure steam (LPS), $0.0674/kWh electricity and $0.0157/t cooling water (Turton et al., 2018). The C_{UT} is estimated by multiplying the unitary cost of each utility with the utility requirements presented in Table 2. The methodology reported by Ulrich and Vasudevan (2004) has been employed to estimate the C_{OL} considering the total number of workers required in the industrial plant based on the annual plant operation (Table 2), the working time of each worker and the average labour cost. The C_{RM} is estimated by multiplying the unitary raw material costs with the raw material requirements. In the case of the Kraft lignin, as dominant raw material, for the economic model has been consider different purchase value were ranged between $350/t and $700/t (Moretti et al., 2021). Additionally, the industrial symbiosis scenario where the designed plant is integrated with the Kraft lignin production plant was evaluated. In this case the kraft lignin can be used directly, without the need of drying, storage and transportation, resulting quite lower kraft lignin price where for this study consider at $100/t. Discounted cash flow (DCF) analysis has been carried out using the parameters reported by Humbird et al. (2011), namely 10 % interest rate, 30 years plant lifetime, 100 % equity financing, 7 years for depreciation based on the Modified Accelerated Cost Recovery System (MARS), 3 years plant construction period and working capital estimation as 5 % of the FCI. The TEA indicators used in this study are the Net Present Value (NPV), the Discounted Payback Period (DPP) and the Minimum Selling Price (MSP).

2.3. Environmental assessment

With the aim of assessing the environmental loads associated with the valorization of lignin for the production of bio-oil and oligomers, the LCA methodology has been applied. It is a well-recognized procedure to determine the environmental profile of biorefinery process, looking for analyze which are the main hotspots of the process, namely, the stage/inputs/outputs that entails the gross environmental impact, and to propose alternative scenarios, besides the identification of the advantages in comparison with its counterparts: fossil-based resources (Arias et al., 2021; Bello et al., 2021; Brancoli and Bolton, 2019; Hetherington et al., 2014). The LCA methodology, according to ISO 14040 and ISO 14044 (Finkbeiner et al., 2006), consisted of four main steps: (1) definition of the goal and scope of the assessment, in which the system boundaries and the functional unit are also defined and selected; (2) life cycle inventory, a key stage in which all process inputs and outputs are collected; (3) impact assessment, to quantify the contribution of each life cycle inventory item to environmental impacts, while also identifying the calculation methodology used; and (4) interpretation of results, which includes identifying hotspots—key contributors to the environmental profile—and conducting sensitivity analyses aimed at enhancing environmental sustainability.

With respect to the goal and scope definition, the base case scenario has been based on considering the functional unit as the recovery process of 1 kg of kraft lignin. The reason behind this selection has been based on the consideration of developing comparison scenarios with other non-renewable resources, as fossil-based. On the other hand, looking for the system boundaries, a cradle-to-gate approach has been considered, as the main focus of this assessment is evaluating the recovery process of lignin for the production of bio-oil and oligomers for further industrial applications.

The life cycle inventory of the production process under evaluation is defined on Table 3. Regarding the database, EcoInvent database has been used to assess the environmental loads. As the EcoInvent database does not include data for Kraft lignin, bibliographic sources have been used to assess its associated environmental loads, applying mass allocation for the overall Kraft pulping process. Notably, a conventional Kraft pulping process produces approximately 1.7 tons of black liquor per ton of pulp (IEA Bioenergy, 2007), leading to a mass allocation of 62.96 % for the black liquor. This analysis uses background data from Bernier et al. (2013) to evaluate the environmental profile of Kraft lignin.

The methodology for determining environmental impacts follows the Environmental Footprint/ILCD recommendations (Commission, 2010). To this end, the impact categories under assessment, together with the method used for its calculation, are depicted on Table 1. Besides, in order to computational implement the inventory data, the SimaPro software has been used (PRé Consultants, 2004). Besides, for assessing the comparison between the scenarios of the bio-adhesives, together with the one of phenol formaldehyde fossil-based adhesive, the ReCiPe EndPoint methodology has been used.

3. Results

3.1. Techno-economic evaluation modelling. Mass and energy balances based on the preliminary techno-economic assessment

Fig. 2A presents the FCI of the whole process considering different plant capacities for the production of both products (BCD-oligomers and BCD-oil). The values were expressed as $/kg and decreased with the increase of the plant capacities, due to the economy of the scale applied in the economic model. The maximum FCI of $2.44/kg and the minimum FCI of $1.03/kg were estimated at an annual production capacity of 10 kt and 100 kt, respectively. It is observed that the changes of FCI values, after the 50 kt annual production capacity, were not significant. As a result, the 50 kt annual production capacity was considered as the optimal plant capacity (OPC) for this process. Table 2 present the FOB purchase equipment cost (Cp), FCI, workers, C_{OL}, utility requirements and C_{UT} at the OPC.

The COM (Fig. 2B) was estimated by taking into consideration the different purchased prices of the kraft lignin ($100-$700/t) as dominant raw material for this process. This approach was conducted in order to determine the sensitivity of the production cost with the price of the raw materials. Fig. 2B illustrates the COM values in $/kg at various plant capacities (10–100 kt/y). At the OPC (50 kt) the COM ranged between of 0.61–1.48 $/kg when the prices of the kraft lignin were $100-$700/t. The COM values seem to have a significant difference based on the different values of raw material which indicates their dependance with the prices of the raw

Table 1
Recommended methodologies for calculating the impact categories values.

Category	Indicator	Methodology
CC	Global warming potential, kg CO_2 eq.	IPCC 2013 model
OD	Ozone depletion potential, kg CFC11 eq	EDIP model
HT,c	Comparative toxic unit for humans, CTU_h	USEtox 2.1 model
AD	Accumulated exceedance (AE), mol H^+ eq	ILCD
ET	Accumulated exceedance (AE), mol N eq	ILCD
EF	Fraction of nutrients reaching freshwater, kg P eq	ReCiPe 2016
EM	Fraction of nutrients reaching marine, kg N eq	ReCiPe 2016
TF	Comparative toxic unit for ecosystems, CTU_e	USEtox 2.1 model
RM	Abiotic resource depletion, kg Sb eq	CML 2002
RF	Abiotic resource depletion-fossil fuels, kg oil eq	ReCiPe 2016

Adapted from Environmental Footprint recommendations Annex 1–2.

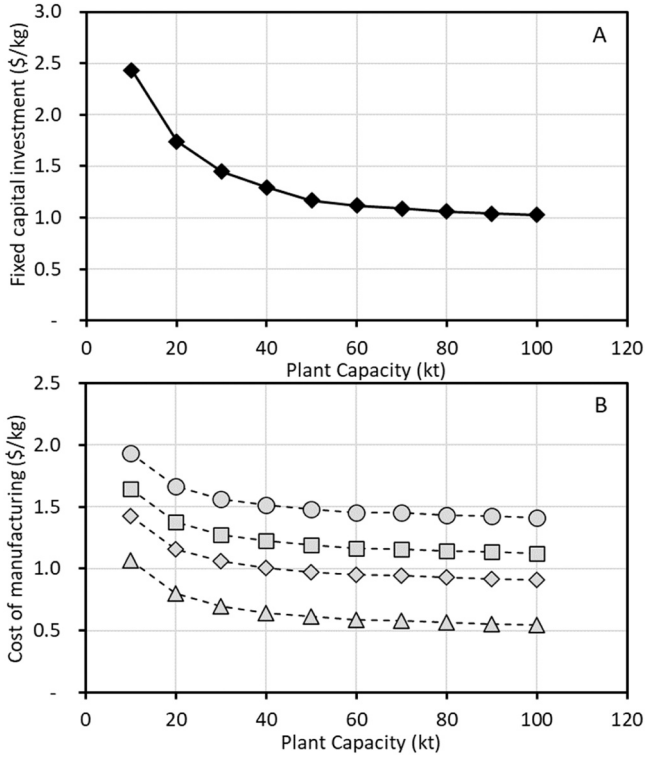


Fig. 2. FSI (A) and COM (B) expressed per kg of the total products (BCD-oligomers and BCD-oil), estimated at varying production capacities (10,000–100,000 t_{tp}/year). The COM estimated taking into consideration different kraft lignin purchase prices (Δ) $100/t, (◇) $350/t, (□) $500/t, (○) $700/t.

Table 2
FOB purchase equipment cost (Cp), FCI, workers, C_{OL}, utility requirements and C_{UT} for 50,000 t/year total products (BCD-oligomers and BCD-oil) production capacity.

Unit		N° Unit	Size (X_t)	Purchase Cost (M$)	Electricity (MWh/y)	Natural gas (kg/y)
V−101	Mixing tank	1	V= 75 m^3	0.20		
A−101	Agitator	1	P = 64 HP	0.12	382.35	
HP−101	High pressure Compressor	1	P = 890 kW	0.384	7052.28	
E−101	Heat exchange	1	A= 236 m^2	0.103		
E−102	Heater	4	Duty= 8 MW	1.276		4864.20
FR−101	Flow reactor	1	V= 20 m^3	0.094		
E−103	Heat exchanger	2	A= 78.6 m^2	0.138		
FV−101	Flash Tank	1	V= 75 m^3	0.200		
V−102	Mixing tank	1	V= 75 m^3	0.200		
A−102	Agitator	1	P = 64 hp	0.12	382.35	
F−101	Filter	1	A= 242 m^2	0.335	94.40	
DR−101	Dryer	1	Q= 1.13 kg/s	0.567	107.56	
EV−101	Evaporator	1	A= 404 m^2	3.807		
C−101	Compressor	1	P = 2331 kW	0.508	18,463.25	
E−104	Heat exchanger	2	A= 553 m^2	0.431		
EXV−101	Liquid-Liquid Exchange Vessel	1	V= 150 m^3	0.154		
DV−101	Decanting Tank	1	V= 36 m^3	0.078		
EV−102	Evaporator	1	A= 11 m^2	0.435		
C−102	Compressor	1	P = 8 kW	0.032	56.04	
E−105	Heat exchanger	1	A= 33.63 m^2	0.034		
DR−102	Dryer	1	Q= 0.28 kg/s	0.430	26.89	
FCI (M$)				**62.30**	**C_{UT} (M$/y) =**	**0.83**

materials.

Regarding the minimum selling price (MSP), it was estimated individually, considering only the FCI and COM of the operating units that are required for the production of each product (BCD-oligomers and BCD-oil). More specifically for the BCD-oligomers MSP

estimations used the operations units from the mixing tank until the first evaporator (EV-101) and dryer (DR-101) (Fig. 1). For the BCD-oil MSP, used all the operating units (except from the DR-101) as illustrated in the process flow diagram of Fig. 1. At the OPC of 50 kt/y total product production, the 72.7 % represent the BCD-oligomers (36.4 kt/y) and the 27.3 % corresponds to BCD-oil (13.6 kt/y). The minimum MSP values at this capacity estimated at $0.65/kg (Fig. 3A) and $0.78/kg (Fig. 3B) for the BCD-oligomers and oil respectively, when the price of the kraft lignin was $100/t. Considering the NPV (Fig. 4) the estimation was carried out using different selling prices for the products and different capacities using the kraft lignin price of $100/kg. The given values represent conservative market prices of fossil-based phenols and ranged between $0.8–1.2 /kg. At the OPC the NPV is positive in all the products market prices. Specifically, with market prices of $0.8/kg for both products the NPV estimated into M$6.5 and DPP at 24 years. This DPP is not acceptable for plant investments (Zuberi et al., 2017), but can be reduced in higher production capacities, until 11 years, at the plant capacity of 100 kt. On the other hand, using the market prices of $1.0/kg and $1.2/kg for both products, at the OPC, the DPP was estimated to 8 and 6.5 years, respectively.

3.2. Environmental assessment

The LCA inventory was based on the energy and mass balances that derived from the process design. The inventory expressed per kilo of the kraft lignin that introduced in the system. The global inventory data used for developing the LCA is depicted on Table 3.

According to the recommended methodologies for calculating the environmental loads associated with the use of lignin as renewable feedstock for obtaining bio-oil and oligomers (Table 1), the following environmental profile has been obtained (Fig. 2).

As it could be seen, the use of chemicals for dissolving the lignin (NaOH) and for the precipitation of lignin oligomers (HCl) are the ones that leads to the higher environmental contribution, being the sodium hydroxide the one with the huge impact, accounting for more than 40 % of the global impact for most of the impact categories under assessment. The energy requirements (both natural gas and electricity) are not an issue in this case, as the biggest impact, as expected, is observed on RF impact category, but is not the main hotspot that could be identified. On the other hand, it should be noted that the use of kraft lignin also entails certain impact over the categories, mostly in the case of HT,c category, contributing for more than a 20 % of the global impact. The reason behind this impact is because lignin has not been considered as a zero-impact resource, as it comes from a background pulping process. The appropriate allocation has been made to assign the impact to the black liquor containing this kraft lignin, in order to bear in mind the environmental loads coming from this stream. But, indeed, in the sensitivity analysis, this kraft lignin will also be considered as a zero-impact stream. As this stream is being avoided to reach the environment without being recovered, significant impacts are prevented, so it could be a good approach to consider this kraft lignin as an "avoided" harmful stream, and this benefit is translated by considered it as a zero-impact input feedstock.

On the other hand, looking for scoring the impact category values, the Table 4 is depicted. It includes the environmental loads associated with the functional unit considered for the study, in this case, the treatment of 1 kg of kraft lignin. As it could be appreciated, the human toxicity harmful of the recovery facility of lignin is not a big deal, in fact, it entails a very low health damage. On the other hand, both OD, EM and RM categories has a very low value, only reaching 0.32 mg CFC_{11} eq, 41.9 mg N eq and 13.4 mg Sb eq per kg of kraft lignin treated, respectively. This could be categorized as a good and sustainable result, as the impact over the quality of ozone layer, eutrophication and mineral resources is not significant. The same conclusion has been achieved when assessing AD and ET categories, as the impact values only reaches mmol quantities. Even though the values for EF and RF categories are a little bit higher, those could also be considered as acceptable scores. However, attention should be paid to the RF value, which amounts to 0.29 kg oil eq/kg kraft lignin used, which is derived from the energy consumption required to produce the different chemicals used in the background process. To reduce this value, different alternatives can be developed, the first of which would be to increase the

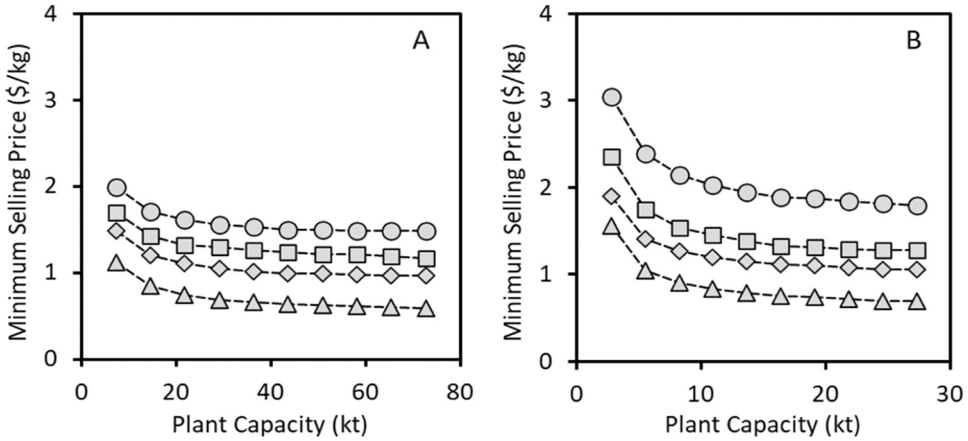


Fig. 3. The MSP expressed per kg of BCD-oligomers (A) and for BCD-oil (B) products. Estimated at varying production capacities (A 7.27–72,700 t/year, B 2.73–27.3 t/year). The MSP estimated taking into consideration different kraft lignin purchase prices (Δ) $100/t, (◊) $350/t, (□) $500/t, (○) $700/t.

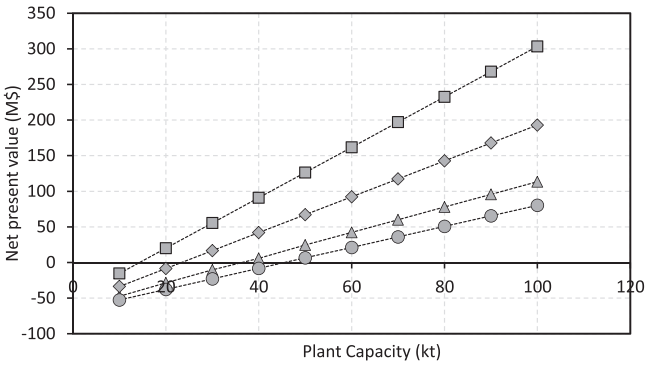


Fig. 4. NPV of the plant at different plant production capacities using different selling prices of the BCD-oligomers and BCD-oils. (○) BCD-oligomers \$0.8/kg and BCD-oil \$0.8/kg, (Δ) BCD-oligomers \$0.8/kg and BCD-oil \$1.0/kg, (◇) BCD-oligomers \$1.0/kg and BCD-oil \$1.0/kg, (□) BCD-oligomers \$1.2/kg and BCD-oil \$1.2/kg.

Table 3
Life Cycle Inventory of the recovery process of kraft lignin to BCD-oligomers and BCD-oil. Functional unit: 1 kg of kraft lignin.

Inputs from technosphere			Outputs to technosphere		
Item	Amount	Unit	Item	Amount	Unit
Kraft lignin	1.00	kg	BCD-oligomers	0.62	kg
NaOH	0.28	kg	BCD-oil	0.23	kg
HCl	0.25	kg			
Energy			Waste to treatment		
Natural gas	0.08	kg	Condensate gaseous by-products	0.15	kg
Electricity	0.25	kWh			

Table 4
Impact values associated to lignin valorization to the production of bio-oil and oligomers. Funtional unit: 1 kg of kraft lignin.

Impact category	Value	Unit	Impact category	Value	Unit
CC	0.83	kg CO_2 eq	EF	0.46	g P eq
OD	$3.22 \cdot 10^{-7}$	kg CFC11 eq	EM	0.04	g N eq
HT,c	$7.77 \cdot 10^{-11}$	CTU_b	TF	$2.79 \cdot 10^{-3}$	CTUe
AD	4.81	mmol H^+ eq	RM	13.4	mg Sb eq
ET	8.09	mmol N eq	RF	0.29	kg oil eq

productivity of the process by improving the efficiency of the lignin treatment technology, thus making a more efficient use of resources. The second option could be focused on the use of renewable energy sources to reduce the consumption of fossil resources. This alternative, although totally viable, and perhaps the most appropriate from the point of view of environmental sustainability, is outside the considered system boundaries, since it depends on the previous processes of raw materials and chemicals. For this reason, the

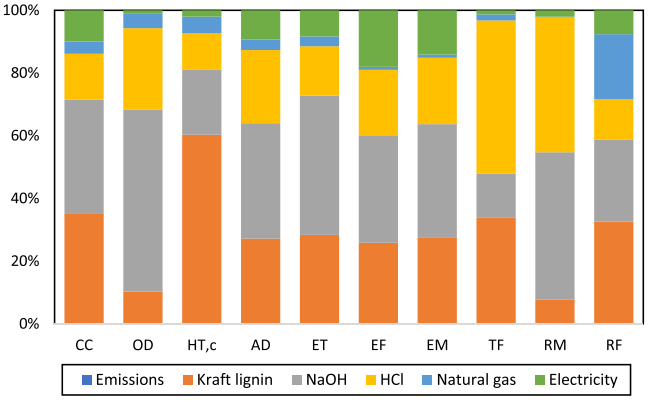


Fig. 5. Environmental profile of the production of bio-oil and oligomers using lignin as renewable input resource.

sensitivity analysis will opt for the use of renewable energies to obtain the energy needed in the process itself, but given their low contribution to this impact category, the improvement in this impact category value would not be significant.

In addition to the environmental assessment, an analysis of both mass and economic allocation for the two products has been conducted to determine which product, BCD-oil or BCD-oligomer, has greater potential from both environmental and economic perspectives. For mass allocation, the production values shown in Table 3 were used (0.62 kg of BCD-oligomers and 0.23 kg of BCD-oil). For economic allocation, as defined in Section 3.1 of this manuscript, average market prices of $1.0/kg for BCD-oligomers and $1.2/kg for BCD-oil were applied. The results from these allocation procedures are presented in Fig. 6.

As observed, when using mass allocation, the environmental impact of BCD-oligomer is significantly higher, as a greater amount of this product is obtained per kilogram of Kraft lignin. However, under economic allocation, BCD-oil has a higher environmental contribution to the profile due to its higher market price. Therefore, the preference for one product over the other depends on the allocation method used; for mass allocation, higher productivity of BCD-oil is preferable due to its lower environmental impact, whereas for economic allocation, increasing BCD-oligomer production is the more sustainable option.

3.3. Sensitivity analysis

In order to evaluate the potentiality of improving the environmental profile and impact loads achieved, sensitivity analysis are proposed, based on the identified hotspots of Fig. 5. Specifically, the following alternative scenarios are assessed, including also the use of renewable energy for the energy requirements, even though is not a major contributor of the environmental profile obtained: (1) Reduce the dose of NaOH by 20 %, (2) use of an alternative base, KOH, (3) reduce the dose of HCl by 20 % and (4) use of renewable energy for energy requirements.

The impact loads obtained are depicted in Fig. 7. As it could be observed, among the sensitivity analysis proposed, the reduction on the NaOH dose by 20 % is the one that provides the highest environmental loads reduction, with an average of 11.29 % with respect to the base case scenario. On the contrary, the alternative of using KOH as base instead of NaOH does not provide improvements on the environmental impacts, but enhancement in some of the impact categories, as the case of EM, CC and RF, with an average of increase that amounts to 29.84 %. With respect to reducing the dose of HCl, the decrease of the environmental loads is lower in comparison with the one of NaOH, in this case with an average value that amounts to 3.94 %, with AD and EM the impact categories with the highest reductions observed, amounting to 5.19 % with respect to the base case scenario. When assessing the use of renewable energy, the highest reduction on the environmental impact is observed in the category of RF, as expected given the avoidance on the use of fossil

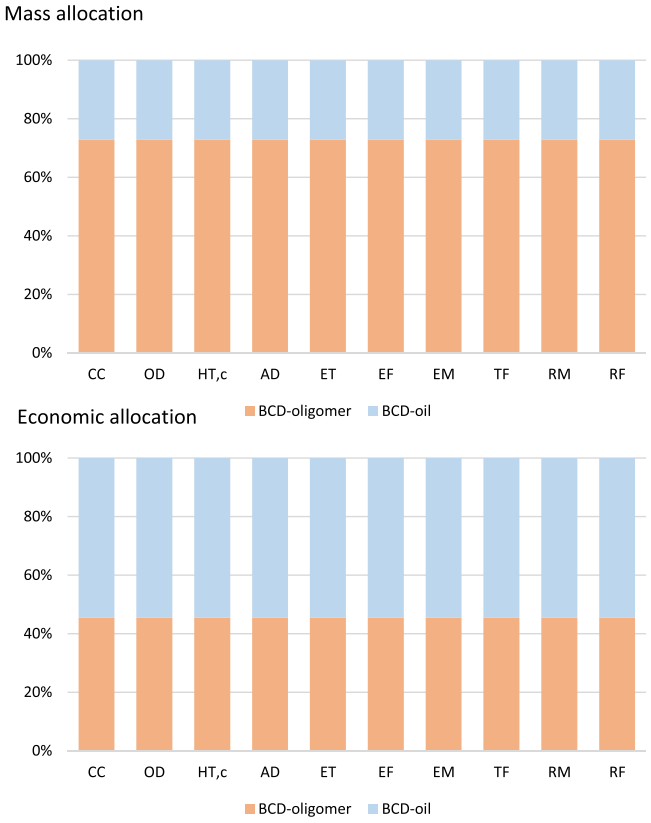


Fig. 6. Environmental profile considering mass and economic allocation.

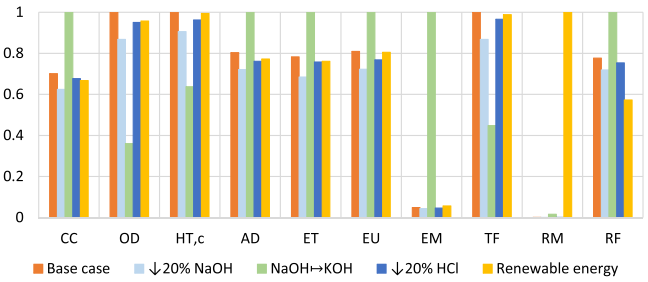


Fig. 7. Sensitivity analysis.

fuels for energy production, but this implies a significant increase on the RM impact category, mostly because of the need for mineral based materials for the construction of the equipment needed for the renewable energy production. But, on average, the use of renewable energy implies a reduction of 5.50 % on the environmental loads.

Given the results obtained it could be stated that the optimization on the use of the chemicals, namely NaOH and HCl, is the best strategy for the improvement of the sustainability potential.

3.4. Combining BCD-oil and oligomers to produce adhesives

One of the most common applications of lignin derivatives is its use for the formulation of adhesives. In this case, it has been proposed and proven the partial substitution of the phenol content on phenol-formaldehyde (PF) resins with the BCD-oil and oligomers. Concretely, according to the report developed by (Solt et al., 2018), the BCD-oligomers could substitute by a 70 % the amount of phenol required in the PF resins, while in the case of BCD-oil, only a 50 % of the phenol could be substituted in order to maintain the tensile shear strength score in the same value to that of PF resins.

To this end, three scenarios are being proposed for assessing the environmental loads and techno-economic scores, looking for determining the benefits and adequacy of the use of lignin oligomers and oil for the production of partial-substitute phenol-formaldehyde adhesives. The environmental assessment has been done using the ReCiPe EndPoint methodology, as it gives three damage categories and a single score value that encompasses the midpoint impact categories previously assessed. However, the functional unit considered for the study has been modified. In this case, the production of 1 kg of adhesive is the one selected, as it is the most widespread when aiming to assess and compare the environmental loads of scenarios having the same purpose, in this case, the production of an adhesive. Besides, the environmental data for the synthetic resin, PF, available on the literature and on EcoInvent database (Wernet et al., 2016), is based on the same approach: the production of 1 kg of adhesive as functional unit. The damage loads obtained when applying the ReCiPe EndPoint methodology are depicted on Table 5.

As it could be seen, in all the damage categories the substitution of the phenol amount implies a reduced environmental impact. However, it should be noted that the production of the BCD-oil and oligomers, even though those are coming from renewable resources, requires the use of chemical agents, that entails certain environmental contribution. The fact of the recovery and use of non-fossil-based resources does not always imply that the impacts over the environmental are lower, as the process of its valorization needs to use chemicals and requires energy, coming from fossil sources. Whereas the damage scores of the alternative proposed scenarios could be considered as a more sustainable option when looking for the partial substitution of PF resins. But the fact that the amount of formaldehyde is the same, which is the one entailing the higher impact over the human health damage category, due to its toxicity and carcinogenic potential, derives on lower improved score on the human toxicity damage category, with a reduced damage of 22 % and 15 % for the oligomer and oil bio-adhesives, respectively (Fig. 8). On both ecosystems and resources damage categories, the use of the PF-BCD-oligomer implies a higher damage reduction in comparison with the PF-BCD-oil bio-adhesive, obtaining a reduced impact value of 53 % and 30 %, respectively. To this end, even though the variations are not as significant between the two proposed scenarios, it could be established that the alternative of PF-BCD-Oligomer adhesive is better, in an environmental point of view, as the impact loads are lower.

Given that the proposed lignin-based bioadhesive relies on preliminary experimental data and has not yet been optimized, there is substantial potential to reduce its environmental impact and enhance its sustainability value. In contrast, phenol-formaldehyde adhesives are already fully optimized at an industrial scale, making further environmental improvements unlikely. These points suggest that the scores obtained for the bio-based adhesive indicate significant potential for advancing sustainability in the adhesives sector. Similar conclusions about the potential of bio-based adhesives have been reached by previous research reports developed by the authors (Arias et al., 2022, 2020).

4. Discussion and conclusion

In summary, it could be determined that the valorization of Kraft lignin to produce bio-oil and bio-oligomers is a sustainable end-of-life alternative. The LCA methodology and the TEA analysis have confirmed the suitability of the process, when a capacity of 40 kton is, at least, considered for the facility. However, with the aim of acquiring better results, future research efforts should focus on the reduction on the dose of chemicals, particularly NaOH, for reducing it contribution over the environmental impact of the production

Table 5
Recipe EndPoint damage loads and single score value of the adhesives scenarios.

Damage category	Unit	PF	PF-BCD-Oligomers	PF-BCD-Oil
Human health	mPt	84.94	66.59	71.88
Ecosystems	mPt	4.03	2.83	3.18
Resources	mPt	7.20	3.38	4.47
Single score	mPt	96.18	72.81	79.53

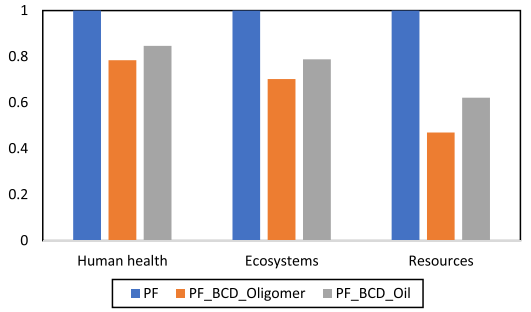


Fig. 8. Comparison of the damage loads for the alternative adhesives scenarios. Acronyms: PF (Phenol-formaldehyde), PF-BCD-Oligomers (substitution of phenol in PF resin by 70 % with lignin BCD-Oligomers) and PF-BCD-Oil (substitution of phenol in PF resin by 50 % with lignin BCD-Oil).

process: the lower the chemicals, the higher sustainability.

Besides, looking for a further use of the co-products obtained by the base-catalyzed depolymerization lignin recovery, BCD-oil and BCD-oligomers, those have been used for the production of bio-adhesives. Although both co-products could be considered as suitable materials to obtain a resin with the appropriate mechanical and physical properties, BCD oligomers seem to be the more suitable option because (1) the amount of BCD oligomers produced is almost 4 times higher compared to BCD oil per kg of kraft lignin being valorized, (2) the damage potential is higher for the BCD-oil product, and (3) although the average selling price of BCD-oil is slightly higher than that of BCD-oligomers, it does not compensate when compared to the production capacity, i.e. it is not 4 times higher, but only 25 % higher. Therefore, these facts imply that BCD-oil is less competitive when production capacity, environmental impact and selling price are taken into account.

In conclusion, bio-based adhesives based on phenol-substitution with BCD-oligomers coming from the valorization of Kraft lignin could be considered as a next generation class of resins that can highly contribute to reduce the environmental impacts associated with the production of synthetic adhesives.

CRediT authorship contribution statement

Ioannidou Sofia Maria: Writing – review & editing. **Ladakis Dimitrios:** Methodology, Investigation. **Arias Calvo Ana:** Writing – original draft, Methodology, Investigation, Formal analysis. **Koutinas Apostolis A.:** Writing – review & editing, Supervision, Conceptualization. **Moreira Maria Teresa:** Writing – review & editing, Supervision. **Feijoo Gumersindo:** Writing – review & editing. **Triantafyllidis Konstantinos:** Writing – review & editing, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper

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Data Availability

Data will be made available on request.

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