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From lignin to market: a technical and economic perspective of reductive depolymerization approaches

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Lignin has grown into one of the main candidates to replace fossil-based resources as it is the largest renewable source of aromatic building blocks. The complex structure of polymeric lignin, however, requires depolymerization to simpler building blocks for the chemical industry. One of the most promising depolymerization approaches is reductive depolymerization of which two process configurations are currently studied in pilot scale installations for upscaling to industrial scale: (i) reductive catalytic fractionation (RCF), and (ii) reductive catalytic depolymerization (RCD). Both technical and techno-economic aspects will be covered within this review, discussing the advantages and challenges of both approaches regarding processing, production costs, product output, and applications. In this regard, RCF benefits from its decreased energy and solvent consumption linked with being a one-step process and delivers a product with a high monomer content (~25–45 wt%). RCD, on the other hand, has the advantage of continuous processing and reduced catalyst fouling and delivers a product that mainly consists of oligomers (<10 wt% monomers). The complete overview of both processes presented here addresses their potential, and can guide future researchers, policy makers and companies to make thoughtful decisions on lignin valorization.

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1. The review discusses the recent advances in reductive lignin depolymerization to produce renewable aromatic building blocks for a variety of applications. The focus lies on the two main approaches of reductive lignin depolymerization: reductive catalytic fractionation (RCF), starting from wood, and reductive catalytic depolymerization (RCD), starting from isolated lignin;
2. The benefits and limitations of both processes are discussed from a technical, techno-economic, and application point of view. This clear comparison can be used by companies, policy makers, and investors to make thoughtful decisions on their sustainable strategies for the future;
3. The availability of a pilot installation for each process emphasizes the industrial potential of both processes. However, each process is still faced with certain limitations, critically assessed within this review, that should be targeted in future research.

1. Introduction

Historically, technological revolutions based on the utilization of fossil resources have driven significant advancements in human civilization. However, over the past century, billions of tons of carbon dioxide have been emitted into the atmosphere,[1] far exceeding the Earth's ecological carrying capacity and triggering global climate issues. Consequently, to achieve a sustainable and controllable carbon cycle, the utilization of renewable biomass carbon resources presents a necessary alternative. Lignocellulose is the most prevalent biomass on earth,[2] mainly consisting of cellulose, hemicellulose and

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lignin. At present, large-scale biorefining of lignocellulose has mainly focused on the utilization of the carbohydrate component, as for instance in pulp and paper industry and production of cellulosic ethanol.[3] However, lignin, as the most abundant aromatic polymer in nature, has high potential as a supplementary resource to partially replace fossil resources for the preparation of aromatic platform chemicals.[4]

Lignin is a complex polymer derived from mostly three monomeric units – *p*-hydroxyphenyl (H), guaiacyl (G) and syringyl (S) – of which relative amounts vary based on many factors such as the plant species, the cell groups, age and environmental growth conditions.[1,5,6] These units are polymerized *via* the formation of phenoxy radicals catalyzed by extracellular oxidases forming various C–C and C–O bonding motifs. Among these native linking motifs, aryl ether bonds are most abundant, within which the β-O-4 motif is predominant.[7] Efficient lignin valorization is hindered by this high heterogeneity of the chemical structure and molecular weight distribution.[8,9] This heterogeneity results in highly diverse properties when lignin is used in the production of chemicals and materials, providing significant challenges in chemical reactions and process development.[10] Nevertheless, selective catalytic depolymerization has been demonstrated as an efficient way to disrupt the complex structure of lignin into smaller and simpler subunits,[8] consequently reducing the heterogeneity of lignin and offering platform chemicals (aromatics) useful for further valorization. In recent years, great efforts have been made in terms of developing different technologies to improve lignin depolymerization, including photocatalysis,[11] electrocatalysis,[12] reductive depolymerization,[13] oxidative depolymerization[14] and acidolysis,[15] *etc*. Among these methods, reductive depolymerization allows for selective catalytic aryl ether C–O bond cleavage under relative mild conditions obtaining high yields of a selected set of phenolic monomers. This review aims to give a clear comparison between the two main reductive depolymerization methods that are considered for upscaling to industrial scale: (i) reductive catalytic fractionation (RCF), starting from lignocellulosic biomass (*e.g.*, wood), and (ii) reductive catalytic depolymerization (RCD), starting from a separated lignin stream (Fig. 1).

While RCF starts with whole biomass as the feedstock, RCD utilizes isolated lignin feedstock which is generated *via* biomass fractionation. The fractionation process breaks down the compact structure of the lignocellulosic biomass to separate the different streams and extract the lignin. In both cases, a mixture of aryl ether C–O and C–C bonds is present in the lignin of which usually only C–O bonds are targeted for reductive depolymerization.[16,17] As indicated in Fig. 2, the average bond dissociation energy (BDE) of C–C bonds is higher than C–O bonds and the retention of aryl ether bonds is thus essential to maximize the yield of phenolic monomers. This concept has been coined as the biorefinery strategy of "lignin-first", which considers upstream lignin structural preservation essential to achieve complete lignocellulose valorization.[18] Notably, this concept includes that value potential is also preserved from the remaining lignocellulose fractions, besides lignin fractions. To achieve efficient lignin-first biorefining using reductive depolymerization, RCF and RCD are seen as the two main strategies upon which various emerging technologies are based.[18–20] Both yield a product oil containing the targeted lignin derived (phenolic) monomers combined with dimers, trimers and oligomers that originate from uncleaved lignin (C–C) bonds. Each of the two strategies has general conceptual advantages and challenges which will be outlined shortly below.

2. Reductive catalytic fractionation

RCF is a lignin-first fractionation method which combines solvolytic lignin extraction with catalytic lignin depolymerization

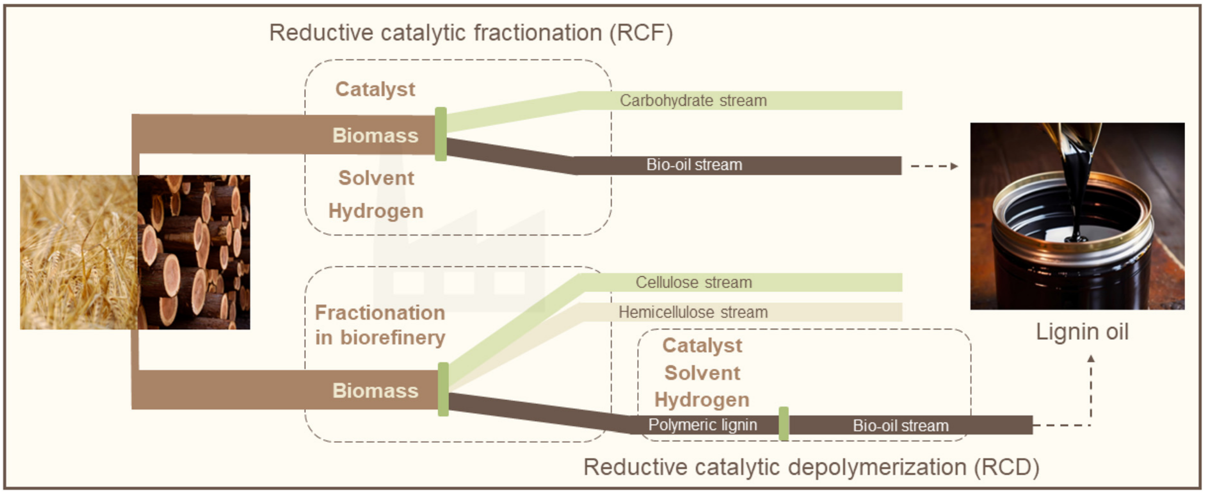
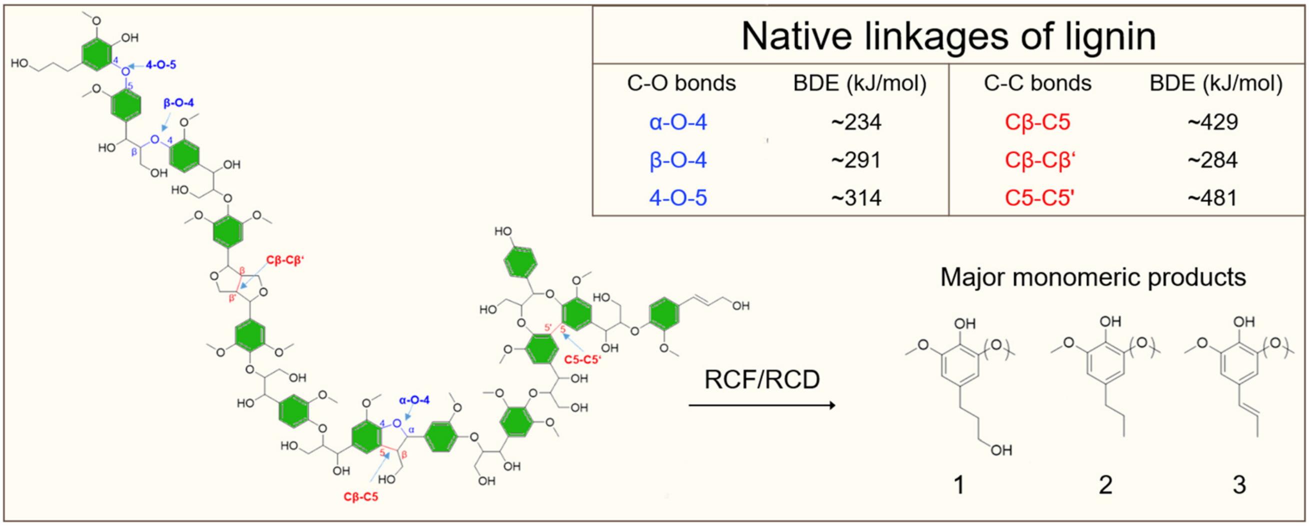


Fig. 1 Two main categories for reductive lignin-first biorefinery strategies, RCF *vs.* RCD, to produce depolymerized bio-oils. While RCF uses a one-step process to produce a bio-oil and carbohydrate stream, RCD uses an isolated polymeric lignin feedstock from a biorefinery fractionation process in which the cellulose and hemicellulose stream have been separated.



Native linkages of lignin

C-O bonds	BDE (kJ/mol)	C-C bonds	BDE (kJ/mol)
α-O-4	~234	Cβ-C5	~429
β-O-4	~291	Cβ-Cβ'	~284
4-O-5	~314	C5-C5'	~481

Fig. 2 Typical C–O and C–C linkages within native lignin from woody biomass, including their bond dissociation energy (BDE)[16,17] and the three major monomeric products formed after depolymerization.

in one process.[18] During the extraction, the lignin is released from the lignocellulose matrix, which is followed by further depolymerization and *in situ* stabilization, which take place simultaneously. RCF can be performed in multiple different reactor configurations (*vide infra*), but are generally performed in batch reactors, which can range from the common laboratory scale 75–300 mL vessels to 50 L reaction vessels.[21,22] Additionally, a 200 L pilot reactor facility at KU Leuven is planned to be used in the future to demonstrate further scalability Fig. 3b.[21]

2.1. Batch RCF

In a batch system, the catalyst (*e.g.*, Ru,[23,24] Ni[21,25] or Pd[24,26]), biomass (*e.g.*, birch,[24] poplar[27,28] or switchgrass[29]), solvent (*e.g.*, methanol or ethanol, often combined with water), and hydrogen (*e.g.*, H_2 or a hydrogen donor)[30] are all placed in a sealed vessel and heated to the reaction temperature Fig. 3a. The solvent facilitates the removal of lignin from the biomass (delignification) of which the extent varies with solvent composition, temperature, and time. The catalyst in RCF serves to intercept and stabilize lignin intermediates in solution *via* hydrogenolysis, hydrogenation, and hydrodeoxygenation reactions.[25,31,32] By maintaining sufficient catalyst availability and hydrogen supply and minimizing the residence time of unstable lignin intermediates in solution, RCF can achieve the maximum monomer yield theoretically possible based on the amount of β-O-4 ether bonds in the native lignin structure as part of the lignocellulose feedstock (theoretical yield). Besides monomers, the RCF oil contains a mixture of dimers, trimers, and larger oligomers due to the presence of C–C bonds in the native lignin structure that are not cleaved in RCF.[33,34] RCF has been demonstrated on noble[23,24,26] and non-noble[21,25,35] catalysts, both capable of achieving theoretical yields of monomers ranging from ~25–60 wt% of the lignin, depending on the biomass feedstock.[18,30]

In this aspect, efforts in multiple directions are made to reduce the economic and environmental impact of the catalyst. A first example is the use of single atom catalysts (*e.g.*, Pd,[36] Ru,[23] Co[35,37]) with reduced catalysts loading (0.18–0.3 wt%) to minimize the amount of metal catalyst added to the reaction.[35] Secondly, the use of earth-abundant metals (*e.g.*, Ni,[21,25] Cu,[38] Co[35,37]) has received substantial attention because of their lower cost and increased sustainability, linked to their higher natural abundance and the reduced environmental impact of their extraction.[39] When comparing the global warming potential (GWP) of the primary production of the refined precious metal Pd (23 700 kg CO_2 per kg Pd)[40] with the refined earth abundant Ni (13.1 kg CO_2 per kg Ni),[41] the huge difference between the two justifies the large efforts that are performed regarding the recycling of catalysts and the transition towards earth-abundant metals. In this regard, bimetallic catalysts (*e.g.*, NiFe,[42] NiCu,[43] RuFeNi[44]) are also gaining interest as these materials can result in synergistic effect to ensure high catalytic performance with improved sustainability and reduced catalyst costs.[45]

In general, the RCF process has two major advantages in comparison to the two-step fractionation + RCD process. Firstly, attributed to the "one batch" configuration of both fractionation and reductive depolymerization, RCF is commonly superb in terms of reducing the reagent consumption such as the solvent as well as energy usage due to a shorter product cycle. The lower reagent and energy consumption could be an important benefit especially when it goes to industrial level. Secondly, lignin condensation during the fractionation is minimized thanks to the immediate stabilization of the active species by the catalyst present, which is difficult to achieve in a typical high-yield lignin isolation process. As a very low number of C–C bonds are created during this process, RCF is the optimal approach to maximize yields of phenolic monomers.[25]

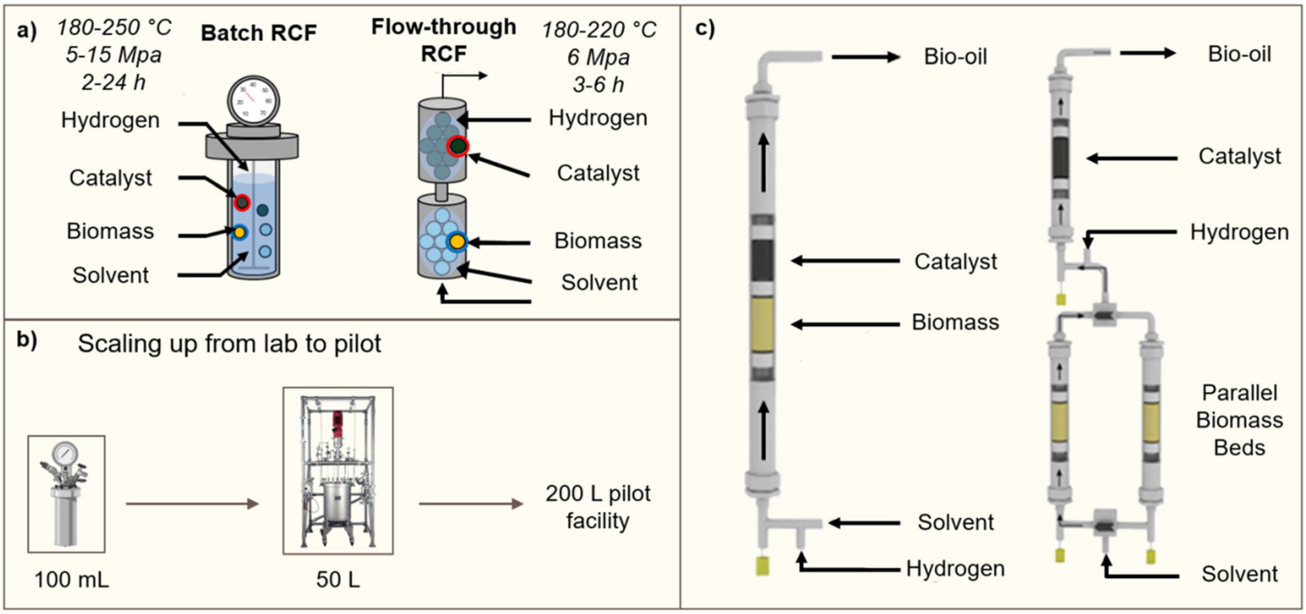


Fig. 3 RCF overview. (a) Overview of the elements of an RCF reaction in either batch or flow-through configuration, including hydrogen (or hydrogen donor), catalyst, biomass, and solvent. (b) Current state of the art with scale up to 200 L batch pilot scale facility in operation at KU Leuven (Belgium).[21] (c) Examples of two flow-through RCF configurations with the biomass and catalyst loaded into the same reactor tube or into separate reactor zones (b was adapted from ref. 21 with permission from the American Chemical Society, Copyright (2023). c was adapted from ref. 32 with permission from Elsevier, Copyright (2017).).

There are also challenges that arise from this one-batch strategy. Firstly, to increase the efficiency of lignin extraction, addition of acid or base are sometimes inevitable, which requires compatibility with the reduction catalyst and can lower carbohydrate retention.[18,46] Secondly, the catalyst requires robustness to other components found in or derived from raw biomass (like organic acid) that can result in catalyst deactivation for example by metal leaching.[25] Another challenge is that in a typical batch RCF system the catalyst remains mixed with the residual cellulose-rich pulp at the end of the reaction. To recover and reuse the catalyst in RCF different approaches have been proposed. Several groups have reported methods to isolate the pulp from the catalyst, including sieving,[25] the use of catalyst pellets which can be removed directly[21,47] or using a bi-phasic solvent combination which has different affinity for biomass and catalyst.[48] Alternatively, the catalyst can be kept separate from the pulp during the reaction using a catalyst basket.[21,25] It is common to find that pellet catalysts have lower yields or require higher loadings than the powder catalysts,[21,25,47] but minimal work has been published on the design and formulation of optimized pellet catalysts or optimized catalyst baskets.

2.2. Flow-through RCF

RCF can also be performed in flow-through reactors. In flow-through RCF, the biomass and the catalyst are loaded into sequential packed-bed reactors Fig. 3. This method is generally categorized as an RCF process as the lignin is not isolated after fractionation but immediately brought into contact with the catalyst. However, as biomass and catalyst are separated in two different reactors, this method can also be categorized as a fractionation + RCD process and has the potential to suffer from some of the disadvantages related to C–C lignin condensation. The biomass and the catalyst can either be back-to-back in the same reactor or loaded into separate heated reactors.[32,49–51] In the upstream bed, the solvent flows through the biomass bed to extract lignin, and then the lignin is carried by the solvent to a downstream catalyst bed for the reductive depolymerization and stabilization reactions. Flow-through RCF provides unique experimental capabilities including independently tuneable solvolysis and reduction temperatures, sequential extraction of different biomass species,[52] and the recovery of isolated pulp and catalyst after reaction for characterization. Flow-through RCF has demonstrated similar lignin extraction and monomer yields as batch RCF.[32,50] While flow-through RCF has continuous flow of solvent and gas, it is not a fully continuous process because the biomass bed must be replaced after extracting the lignin. Flow-through RCF can be operated semi-continuously with a dual-bed system, in which lignin is extracted from one biomass bed while the other can be re-filled. This was demonstrated in a catalyst stability study in which four packed biomass beds were extracted and reduced over a single packed catalyst bed.[32] Fully continuous RCF systems have not been demonstrated to our knowledge. Solids handling across large temperature and pressure ranges represents the major challenge to be overcome for continuous operation. Taking inspiration from the industrial biorefining strategies, reactive screw extrusion or lockhopper continuous stirred tank reactors can be explored in the future as new routes toward fully continuous RCF.

3. Fractionation + reductive catalytic depolymerization

While RCF uses a one-batch process in which all steps (*i.e.* extraction, depolymerization and stabilization) occur in one reactor (Fig. 4a), RCD fully decouples the fractionation and depolymerization steps. As a consequence, new opportunities for optimizing lignin extraction efficiency and limiting catalyst deactivation are provided. The core challenge in the RCD strategy lies in attaining high yield of lignin isolation with well-preserved, aryl–ether C–O bonds, providing a suitable feed for downstream RCD.

3.1. Fractionation of suitable RCD feedstocks

Approaches for lignin isolation suitable for RCD, can be classified in two ways: (i) isolating unmodified (native-like) lignin, which can retain its regular linking motifs (Fig. 4b),[54] or (ii) isolating chemically modified (stabilized) lignin, in which a reagent is used to stabilize the active lignin intermediates as a way to retain the native C–O bonded polymeric structure

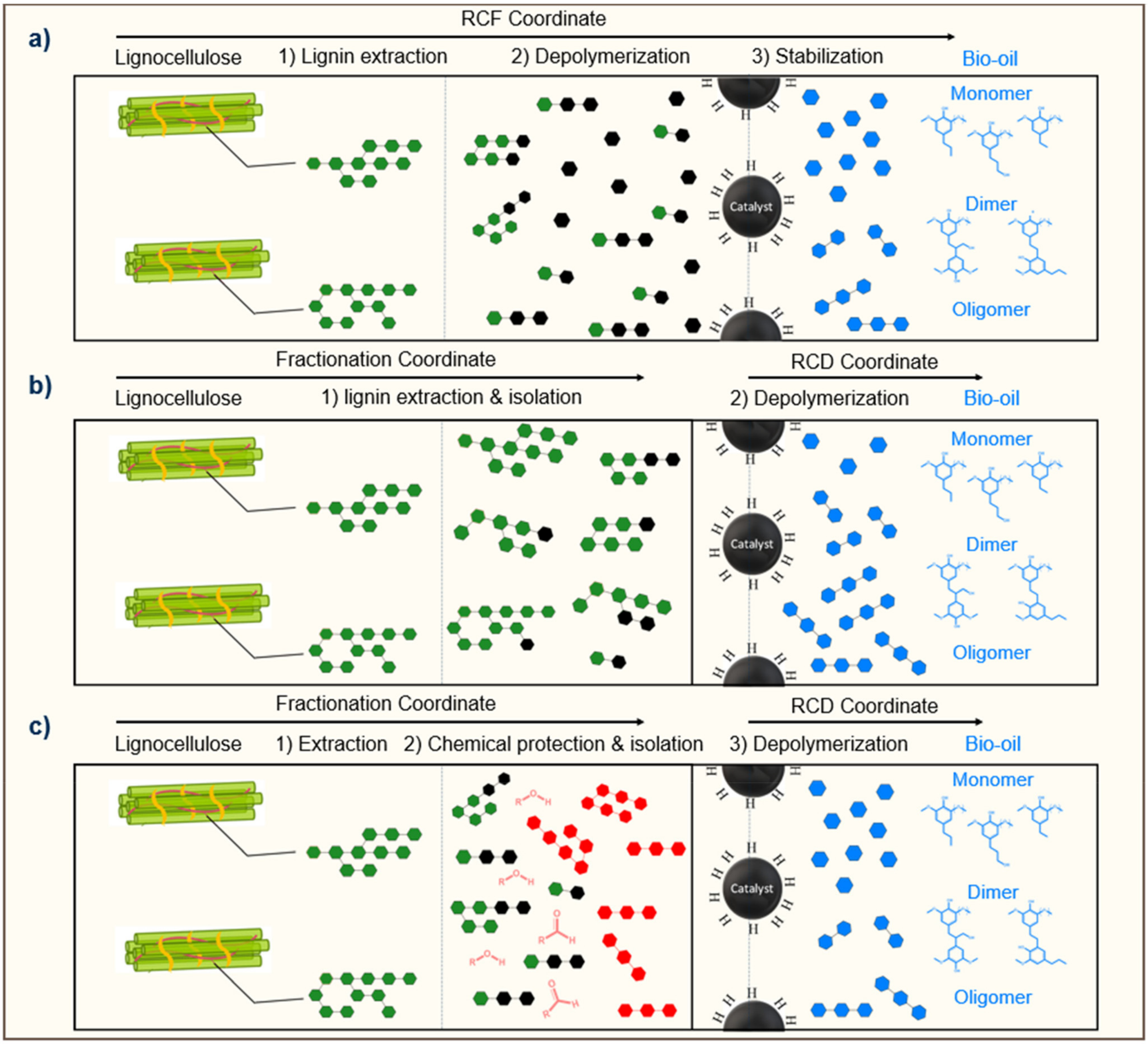


Fig. 4 Schematical representation of (a) RCF coordinate, and (b & c) Fractionation and RCD coordinates in respectively the absence and presence of modifying agents. 🟢 represent native lignin units; ⚫ represent active lignin units; 🔴 represent chemically modified lignin units; 🔵 represent stabilized (depolymerized) lignin units. The modified pathway uses a mechanism of aldehyde/alcohol protection to prevent lignin condensation. The term coordinate is used to represent the progress of the reaction. (Fig. 4 was adapted from ref. 53 with permission from Elsevier, Copyright (2019).)

(Fig. 4c).[20] In this figure, the aromatic units present in the lignin are shown in different colors depending on the state in which they are at that moment within the process. The green units are used to show the native lignin units, which are present in the lignocellulosic biomass and can be extracted when using mild extraction methods, for example milled wood lignin (MWL) extraction. The black units are active lignin units which are produced during the depolymerization and are prone to repolymerization. To inhibit repolymerization, these active units can be stabilized immediately after their generation *via* catalytic hydrogenation (blue units) as is done in RCF (Fig. 4a). Another possibility is the addition of protecting groups which prevents repolymerization by the production of a chemically modified lignin (red units) (Fig. 4c). As this chemically modified lignin still has long chains, a further depolymerization step in presence of a catalyst (RCD) should be performed. In both cases, the bio-oil will contain a high monomer content as repolymerization has been limited. For most lignin fractionation methods, no stabilization step is performed before the isolation which leads to a more condensed lignin. When these more condensed lignins (*e.g.*, technical lignins such as Kraft lignin) are depolymerized in RCD, a product with a higher oligomer content will be obtained (Fig. 4b).

3.1.1. Unmodified pathways. On the other hand, there are also examples from unmodified pathways that are able to obtain native-like lignin. However, the reality is that the smaller the change to the native structure of lignin, the lower the isolated lignin yield that can typically be achieved.[54] For instance, MWL isolated by extensive milling of biomass followed by neutral 1,4-dioxane extraction represents well the native lignin structure but at very low yield due to low extraction efficiency.[55] Despite the conflict between extraction efficiency and lignin quality, there are still some extractions under mild conditions being exploited aiming for the isolation of lignin with a well-preserved unmodified aryl–alkyl ether structure in high yield. For example, Chen *et al.* used an alkaline 1,4-dioxane aqueous mixture (1,4-dioxane : water, 80 : 20, containing 0.05 M NaOH) to extract ball-milled eucalyptus.[56] The yield (29.7%) and purity (71.1% β-O-4 content) of lignin recovered from this extraction are much higher than that of a typical MWL. The application of ionic liquid[57] and deep eutectic solvents[58] as lignin extraction media has attracted increased attention in this context. These extractions indicate the potential to offer high yield native-like lignin suitable for downstream RCD processing but still require many further efforts to make them compatible with RCD at higher TRL due to costly reagent use or complex downstream processing. As an alternative, 'flow-through' lignin extraction seems to be a promising technology for the isolation of unmodified lignin.[32,50,59] In general, the recalcitrance and condensation of lignin largely depends on the residence time of the lignin fragments in the pulp and liquid during the fractionation process.[54] In view of this, Beckham *et al.* developed a flow-through system that enabled the isolation of native-like lignin in a 63.1% yield.[59] Through the continuous flow system, lignin fragments are rapidly removed from the *in situ* reaction system and are quickly cooled down to prevent condensation reactions. The success of this approach was shown by the similar monomer yield that was obtained from the isolated lignin compared to the native biomass.

3.1.2. Modified pathways. Over the past decades, researchers have also developed extraction technologies from the aspect of *in situ* chemical modification/protection, which seem to be alternatives to overcome the challenge arising from the inherent correlation between lignin isolation yield and retention of the native C–O bonded structure stated above. These technologies aim to trap or prevent the formation of reactive species that are formed during extraction preventing condensation and leading to lignin with a modified structure (Fig. 4b). The group of Luterbacher used aldehydes to react with the α and γ alcohols in the β-O-4 motif of lignin to form a cyclic acetal structure, thereby preventing the formation of benzyl carbocations and subsequent C–C condensation (Fig. 4b).[20] This method, known as aldehyde-assisted fractionation (AAF), isolates a modified lignin (73% yield) with a high β-O-4 content (up to 68%) suitable for downstream RCD.[20,60] Alternatively, fractionation with alcohol incorporation has been exploited. Lancefield *et al.* employed various alcohols during high-acid/low-temperature organosolv to initiate a nucleophilic attack on the generated β-O-4 benzylic carbocations to produce α-alkoxylated lignin (Fig. 4b), allowing isolation of lignins in good yields (84 wt% from beech, 97 wt% from walnut shell).[19] HSQC NMR analysis on the isolated walnut shell lignin showed high retention of β-aryl ethers (52%) with a high level of modification of the linkage with butanol (49%). Deuss *et al.* found that this approach not only effectively prevents lignin condensation but also enhances the solubility of larger lignin fragments in solvents.[61]

One disadvantage of the use of modified lignins is that the modifying agent is usually sacrificed during hydrogenolysis. For instance, Luterbacher *et al.* found the formaldehyde used in AAF process will be transformed to methane after hydrogenolysis.[20] Analogously, the alcohol protecting groups will likely be transformed to alkanes during hydrogenolysis consuming additional hydrogen, even though this has not been fully investigated yet. These aspects may be a problem that cannot be ignored from techno-economic perspective, especially when this process is applied on large scale.

3.2. Reductive catalytic depolymerization

After isolation of the lignin, depolymerization is necessary to achieve platform aromatics for further applications. Similarly to RCF, the main part of the research in RCD has been performed in batch reactors using heterogeneous catalysts (both noble and transition metal-based) in which significant progress has been made regarding the optimization of process conditions and the transition towards the more sustainable and less costly earth-abundant metal-based catalysts.[42,64–66] While batch reactors are favored for a fast screening of different process conditions, continuous set-ups are preferred at industrial scale because of the reduced processing times and energy consumption (Fig. 5).[67,68] One of the advantages of RCD is the availability of full-continuous set-ups in which near-theoretical monomer yields can be obtained at the laboratory scale, for example 45% monomer yield compared to 50% theoretical yield for AAF lignin (180 °C, Ni/C, methanol/dioxane 8/2) and 28.4% monomer yield compared to 31.2% theoretical yield for copper-catalyzed alkaline hydrogen peroxide lignin (190 °C, Pd/C, dioxane/water 9/1).[69,70] For technical lignins, continuous systems allowed even better results in comparison to batch systems as the longer contact times between reactants and products in batch systems lead to increased repolymerization and catalyst deactivation.[62] In addition to the possibility of using full-continuous systems, separation of fractionation and reductive depolymerization in RCD avoids interaction between the catalyst and the biomass pulp, which facilitates recycling and extends the catalyst lifetime, like the use of a catalyst basket in RCF.[25] Recently, Jusner *et al.* demonstrated a continuous RCD process in which a stable output of depolymerized product (80 wt% yield) in terms of molar mass, OH-group distributions, and monomer content could be obtained over 77 h of operational time (hydrolysis lignin, 200 °C, Pd/Al_2O_3, methanol).[71] Moreover, a pilot plant at VITO (Belgium), which can produce more than 100 kg of depolymerized lignin per day, is in place to demonstrate further scalability of this process (Fig. 5b).[63]

In addition to these advantages, several challenges also need to be considered for the upscaling of RCD to an industrial scale. The main challenge is the influence of the fractionation method on the feedstock cost, process performance and product quality of the lignin.[72] As the variability in C–O and C–C bonds influences the efficiency of RCD, a high number of aryl ether C–O bonds is a crucial structural feature of the isolated lignin. Most of the current large-scale biorefinery processes produce more condensed lignins rich in C–C bonds (technical lignins), which reduce the efficiency of the depolymerization process and produce high Mw oligomers with a low monomer content.[71] On the other hand, Shuai *et al.* showed that improving the fractionation method (*i.e.*, AAF) significantly enhances the monomer yield (>45%) in comparison to a more condensed, non-protected lignin (<10%) using the same reaction conditions (200 °C, 6 h, THF).[20] To improve the efficiency of the depolymerization of more condensed technical lignins, more research is required to cleave the C–C bonds in a mild and efficient manner.[73]

The employed fractionation conditions will also have a significant influence on the solubility of the lignin which can be challenging for continuous RCD. While the solubility is of minor importance to further processing in batch units, as the lignin is added as a powder, full continuous RCD can only use dissolved lignin to prevent clogging of the reactor bed. Consequently, the undissolved, higher molecular weight lignin cannot be valorized through RCD and can at the moment only be used in lower-value applications, lowering the cost-efficiency of the process.[71] Moreover, the high molecular weight fraction of lignins often contains a high number of β-O-4 linkages which could easily be cleaved in RCD.[74] It is thus key to increase the solubility of these fractions to enhance the efficiency of the RCD process.

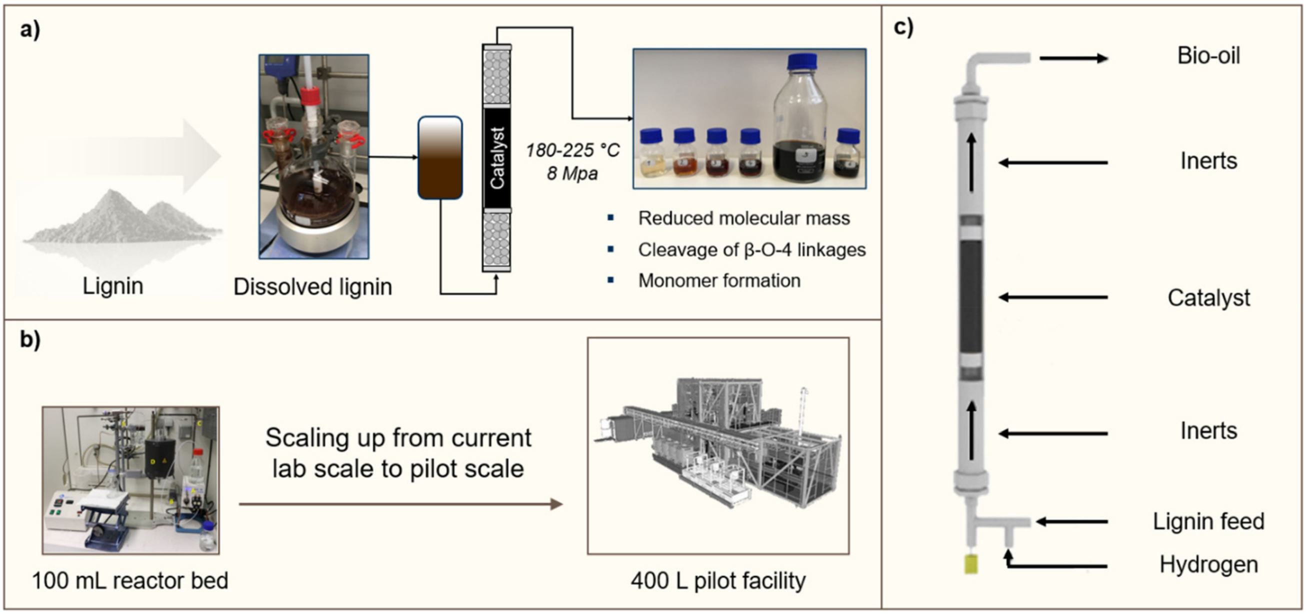


Fig. 5 RCD overview. (a) General overview of the RCD process starting from a lignin powder. The lignin is dissolved which is streamed as feed over a fixed bed reactor to depolymerize the polymeric lignin to monomers, dimers, trimers, and oligomers. (b) Current state of the art scaling up to a 400 L continuous pilot scale facility in operation at VITO (Belgium).[62,63] (c) Zoomed in figure showing the composition and streams present in the fixed bed reactor of the RCD process (b was adapted from ref. 62 with permission from the Royal Society of Chemistry, Copyright (2021). c was adapted from ref. 32 with permission from Elsevier, Copyright (2017).).

Thirdly, the fractionation conditions can affect the performance of the catalyst in RCD by using incompatible reagents during the fractionation step. Examples of the effects of these impurities include a shift in selectivity in presence of chlorine impurities[75] or catalyst deactivation due to accumulation of inorganic impurities on the catalyst.[76,77] Besides these catalyst poisons, several other deactivation methods can be expected depending on the process conditions, such as sintering or leaching. In the full-continuous RCD experiments that are currently available, fouling of organic impurities and support degradation have been observed as the two main catalyst deactivation mechanisms.[71,78] However, more experiments with different catalysts and lignin feedstocks are required to gain a more profound insight on the different mechanisms of catalyst deactivation. In addition to reported recycling techniques (*e.g.*, calcining[70]), a deeper insight into catalyst deactivation will inform efforts in extending the catalyst lifetime, especially for lignin feedstocks that contain significant amounts of impurities. Establishing mild, clean and efficient fractionation protocols together with optimization of the reaction conditions will be key to ensure a technically feasible industrial process.

4. Electrochemical reductive depolymerization

Electrochemical depolymerization of lignin is mainly known under oxidative conditions in which C–C and C–O bonds are broken to yield smaller oligomers and monomers such as vanillin and vanillic acid.[79] While no profound analysis of electrochemical oxidative depolymerization approaches will be given as it falls beyond the scope of this review, the relatively unexplored domain of electrochemical reductive lignin depolymerization will be summarized shortly to give a complete overview of reductive lignin depolymerization methods.

In electrochemical reductive depolymerization, the lignin is cleaved in smaller units by hydrogenolysis of the C–O–C bonds. There are several advantages coupled with this approach. The use of electric potential energy to overcome the activation barriers allows this method to be performed at room temperature and ambient pressures using renewable energy sources, safe and sustainable solvents (*e.g.*, water), and non-noble metal catalysts (*e.g.*, Ni).[80,81] The hydrogen required to perform the reduction step is generated *in situ* as reactive *H species from water electrolysis, removing the pressurized hydrogen required in RCF or RCD.[82] Additionally, the low reaction temperature suppresses undesirable side reactions such as repolymerization.

There are also challenges that still need to be solved to expand electrochemical reductive depolymerization to an industrial scale. One example is the low lignin concentration used during electrochemical reductive depolymerization (<5 g L^{-1})[83–87] which would lead to extensive solvent usage and high process costs at industrial level. The lignin dissolution could be improved by using basic media or other solvents, like ionic liquids and deep eutectic solvents, which are receiving increased attention because of their lower volatility, improved lignin solubility, wider electrochemical window, and stronger interactions between lignin and catalysts.

Another challenge of electrochemical reductive depolymerization is overhydrogenation.[88] To retain aromaticity, hydrogenolysis must be faster than hydrogenation which can be achieved by modulating the adsorption strength and configuration of reactants and intermediates.[82] Zhai *et al.* showed that by adjusting the acidity of the anolyte, the degree of hydrogenation could be adjusted between ketone and alcohol product without compromising the aromaticity.[86] Another recent work by Lindenbeck *et al.* showed that the selectivity could also be directed entirely in the other direction by selective production of aliphatic molecules using a carbon electrode.[85] These examples show that significant hydrogenation in electrochemical reductive depolymerization can be adjusted by altering the reaction conditions to produce the targeted compounds.

A final challenge is the limited number of experiments performed on real lignin substrates. As groups are only starting to explore the domain of electrochemical reductive depolymerization, their efforts mainly focused on understanding its mechanisms using model compounds. The group of Slabon, however, reported multiple works on the use of electrochemical reductive depolymerization using real lignin substrates.[83–85,87] Within these publications, successful depolymerization of Soda and Kraft lignin was observed by reduction of the solution's brown color and formation of phenolic monomers.[83,84] While these works show the potential of working with real lignin, more studies should be performed to investigate the efficiency of the depolymerization and the composition of the product.

Because of the limited information that is currently available on the techno-economic parameters of electrochemical reductive depolymerization. This depolymerization strategy will not be discussed in detail in sections 5 and 6. While more profound studies on this strategy should be conducted to investigate its suitability for industrial scale, the advantages of electrochemical reductive depolymerization show its potential as an alternative to RCF and RCD in the long-term.

5. Techno-economic perspective

While both approaches (*i.e.*, RCD and RCF) have their technical advantages and disadvantages, a techno-economic analysis (TEA) is essential to get insights in the economic feasibility of both processes on an industrial scale.[26,71,89] In this work, a TEA has been conducted for both processes by harmonizing techno-economic data from two recent publications (Table S1, SI). On the RCF front, Arts *et al.* describe how the cost of lignin oil production can be reduced by using a percentage of the product stream as recycled solvents instead of purified solvents.[26] As lowering production costs of depolymerized lignin oil is vital for its industrial application, this article was chosen as the basis for the RCF TEA. On the RCD side, Jusner *et al.*

recently demonstrated a strategy for upscaling the continuous production of RCD-lignin oil using commercially available catalyst, and this article was therefore chosen as the basis for the RCD calculations.[71] In this case, the costs and revenues of the lignin fractionation are considered within the lignin feedstock price. To enable a fair comparison, both processes were scaled to an output of 30 000 ton per year bio-oil production, as this scale is comparable to that used in the original articles, and also in line with feedstock availability and potential economies of scale.[26] Economic parameters like plant lifetime, cost of capital, *etc*. were also kept the same for both processes, and all costs were calculated on a 2023 basis. Additionally, to make the system more generalizable and easier to compare, we have not considered a combined heat and power (CHP) system for either process, making some of the RCF costs differ from those in the original article. Distillation energy requirements have been estimated based on numbers from ref. 71. Further details are available in the SI.

5.1. TEA RCF

For RCF and RCD, two scenarios are analyzed. The RCF base case uses methanol as solvent and does not take recycling of the product stream into account (only recycling of the solvent itself). In this case, solvent and steam (process heat) are the most significant contributors to the overall cost per ton of depolymerized lignin (Fig. 6a). For the improved RCF case, which considers 80% of reaction liquor recycling, the major cost contributors are Capital Expenditure (CAPEX) and feedstock (Fig. 6b). In both RCF scenarios, large revenues are received within the RCF process from the valorization of sugars which significantly reduces the minimum selling price of the depolymerized lignin product. The bio-oil cost of production of this process is €2874 per ton. Considering a 25% tax rate, the minimum selling price for an internal rate of return (IRR) of 10% is €3234 per ton. A possible solution to improve the minimum selling price to more feasible prices for the industry is to implement recycling of the product stream. If a liquor recycling rate of 80% (the highest considered feasible using experimental feedback[26]) is used, the bio-oil cost of production reduces to €1264 per ton with a minimum selling price of €1544 per ton for an IRR of 10%. The main reasons for this sharp decrease are the decreased solvent and steam costs, the former due to the lower solvent flow rate needed, and the latter because of the lower distillation requirements.

5.2. TEA RCD

In the case of RCD, the cost of bio-oil production in the base case scenario is €3459 per ton using assumptions based on the current proven research, which gives a minimum selling price of €4046 per ton for an IRR of 10% (Fig. 6a). The base case cost of production is thus lower for RCF than for RCD. However, significant decreases can be expected in RCD costs upon further optimization of the process. Solubility and solid loading are two key parameters that can be addressed by, *e.g.*, improving the solubilization procedure or the lignin type used. This optimization was shown in an improved model which reduced the bio-oil production price to €1659 per ton and the minimum selling price to €1893 per ton (Fig. 6b).[71] The major cost contributors in RCD are similar to RCF, with the main four items being feedstock, solvent, CAPEX and steam costs.

5.3. Assumptions TEA

In the case of RCF, there is a small variability in price depending on the biomass used to produce the bio-oil. Arts *et al.* showed for example that softwood spruce required a higher minimum selling price than hardwood birch due to the lower lignin extraction from the softwood matrix.[26] However, the potential variability for feedstock cost in RCD is significantly

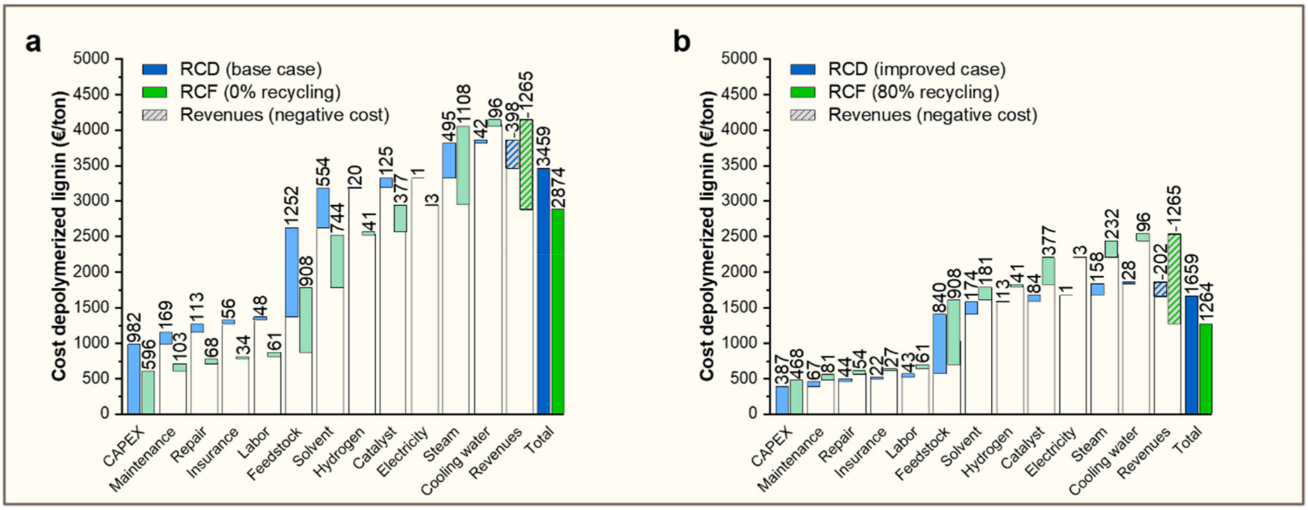


Fig. 6 Cost-breakdown of the different factors determining the price of depolymerized lignin produced *via* RCF (green) and RCD (blue). For each process, two cases are presented: (a) a case currently proven in research (0% recycling/base case) and (b) a case taking a realistically improved scenario (80% recycling/improved case). Valorization of the sugar streams in the RCD case is considered within the feedstock price.

higher due to the different lignin isolation processes that are available. Hydrolysis lignin is considerably more expensive than wood as a feedstock and is only partially soluble in methanol. The partial solubility increases the feedstock demand per unit of bio-oil produced and raises feedstock costs further. Nevertheless, a variety of suitable lignin streams is available for RCD. In general, a distinction should be made between cheaper technical lignin, such as hydrolysis lignin, that contains a higher amount of C–C bonds and more expensive native-like or protected lignin, with a high aryl ether C–O bond content. The choice of a more expensive lignin could still lower the minimum selling price as it could increase the lignin solubility and thus valorize more of the lignin feedstock and decrease solvent costs. For example, organosolv lignin represents an intermediate between hydrolysis and native-like protected lignin in terms of cost but can realize high solubilization yields in organic solvents (up to 100% in methanol or acetone).[90] A final aspect regarding the feedstock price that should be considered is the maturity of the lignin producers and the available volumes on the market. As the biorefinery market is still developing and the valorization of byproducts is yet to be optimized, it is difficult to predict the common lignin feedstock price in the future.

Another difference between the two processes is the solvent consumption. As mentioned above, Arts *et al.* showed an optimized version of the RCF process in which a large amount of the product stream is immediately recycled and used as process solvent, which reduces solvent costs, CAPEX and steam costs. For the RCD study, similar recycling processes are not yet considered and an additional reduction of the minimum selling price of the product could be expected in line with those seen for RCF. In the TEA, a solvent loss of 0.1 wt% is considered for both RCF and RCD which can be caused by alkoxylation or other side reactions involving the solvent under the reaction conditions. Additionally, in the case of RCF, a part of the solvent could get trapped within the biomass pulp during the fractionation.

A third assumption that should be addressed is the choice and lifetime of the catalyst. The higher catalyst costs of RCF in comparison to RCD can be explained by the higher Pd loading (5 wt% Pd on carbon) that were used with regard to RCD (1 wt% Pd on alumina). In both cases, a lifetime of 1 month is considered as more research is required to validate longer catalyst lifetimes at industrial scale.[32,71,78] In this aspect, it will be essential for future studies to evaluate the relative rate of reversible (*e.g.*, poisoning, coking or oxidation) and irreversible (*e.g.*, leaching or sintering) deactivation mechanisms through more extensive time-on-stream and cycling experiments. Some studies have already shown the possibility to regenerate the catalyst after the reaction to prolong its lifetime.[25,70] However, further research is required to enhance the catalyst lifetime and limit the operational time lost due to catalyst replacement.

An important parameter that determines whether the minimal selling price is acceptable depends on the product output of the process, which is of course essential for the targeted applications. While both outputs consist of a mixture of monomeric, dimeric, and oligomeric alkylphenols, the ratio of monomers to oligomers in the product varies significantly. As RCF uses depolymerization of native lignin, less C–C bonds are present and a high monomer content (~25–45 wt%) is obtained in the product.[25,26] While some isolation processes in the RCD approach (*e.g.*, AAF) give similar monomer values in the product to RCF (up to 50 wt%),[70] most fractionation methods lead to lower monomer contents (<10%) and a large oligomeric fraction after RCD.[62,71] In this regard, a low monomer content and large oligomeric fraction is assumed to be a general property of the current RCD product. It is expected that the monomer content in the product after RCD can significantly increase, and become similar to RCF values, once isolated lignin with high ether bond content becomes commercially available.[91] The availability of high ether bond lignin would increase the flexibility of the RCD process as a broad range of products with different monomer and oligomer contents could be attained. As the product compositions of the current RCF and RCD processes differ significantly, the potential applications and consequently the acceptable price range for the products are also likely to differ.

6. Applications

As reductive depolymerization technologies advance, efficiently utilizing the phenolic products has become essential for the economic viability and ultimate success of these methods. The products of lignin depolymerization—reflecting the complexity of lignin itself—typically consist of a mix of monomers, dimers, and larger oligomers. The proportions of these groups depend on the effectiveness of the catalytic depolymerization process, which in turn influences the complexity of the mixture and its ease of separation. Mixtures with fewer, more chemically and physically similar components are likely more suitable for applications in fine chemicals. Conversely, highly variable product mixtures that cannot be easily separated may be better suited for applications where specific physical or chemical properties are more important than purity, such as in solvents, printing inks,[92] or polymer applications like polyurethanes[93,94] and epoxy resins.[22,95] Additionally, chemically modifying the product mixture to adjust its functional properties—or in the case of bulk chemicals, defunctionalizing it—presents multiple opportunities to broaden the range of valuable lignin-derived compounds.

6.1. Platform chemicals

The alkyl phenolic monomers resulting from the reductive depolymerization of lignin feature various functional groups, including *o*-methoxy groups and *p*-sidechains (Fig. 7). These compounds, with some modifications (*e.g.*, amination), show promise as base chemicals for pharmaceutical and chemical uses. In this regard, bio-oil produced in RCF processes is currently better suited for use in platform chemicals because of the higher monomer yields that are obtained. However,

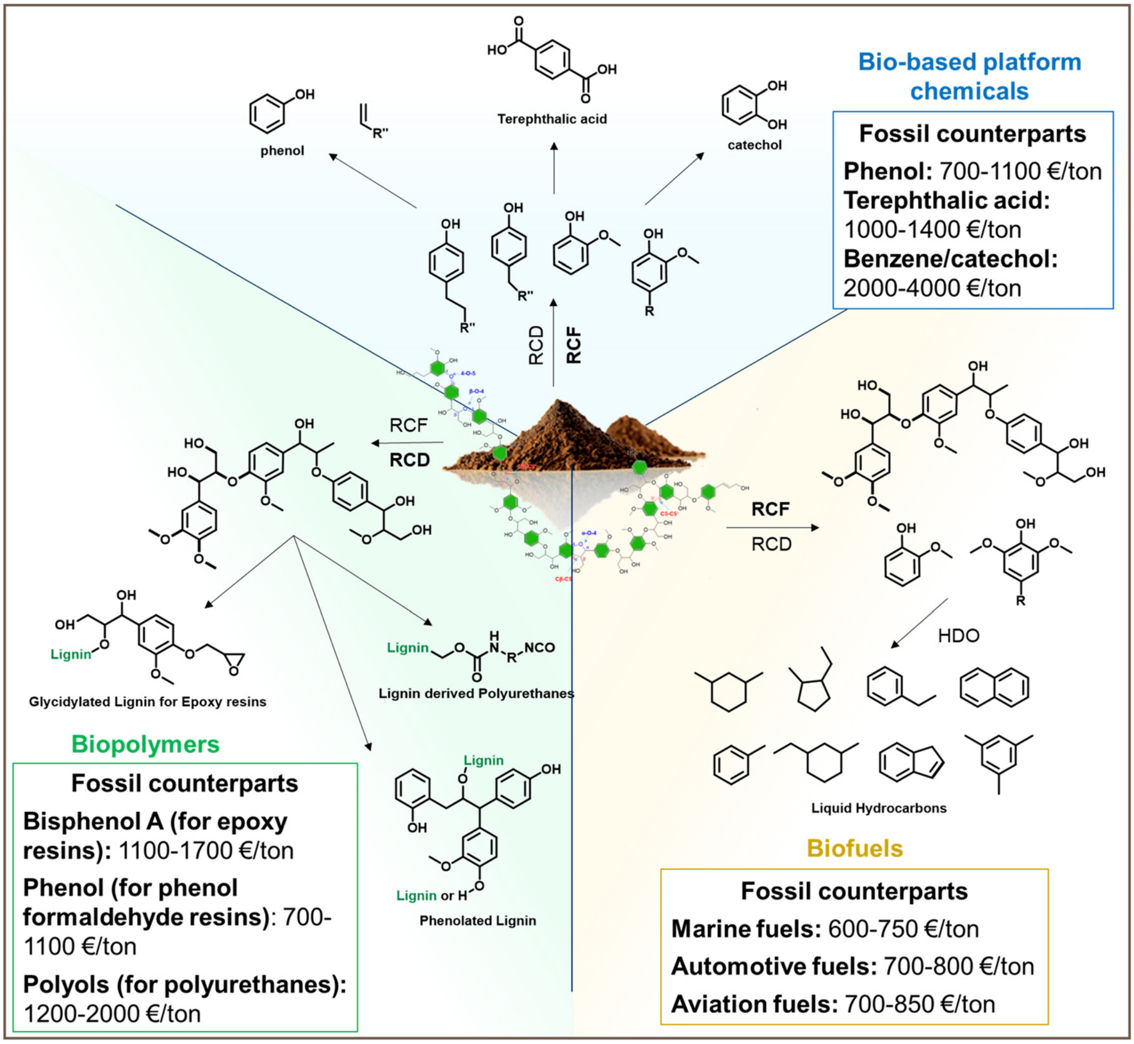


Fig. 7 Different valorization pathways of lignin depolymerization products sourced from RCF and RCD within bio-based chemical, fuel, and polymer applications. Biofuel and bio-based chemical applications are favored for the current RCF process because of the high monomer content reached in the product. Polymer applications, on the other hand, are better suited for the smaller oligomers that are mainly produced *via* the RCD process.

despite its potential, obtaining sufficient selectivity for one type of monomer in the bio-oil mixture remains challenging.

The composition of the product mixture from lignin or biomass depolymerization can be tailored based on the specific process conditions, catalyst selection, and type of biomass used. For instance, using Ru/C as a catalyst in the RCF of biomass yields 75% selectivity for 4-propyl phenolics, while Pd/C promotes the formation of propanol-substituted phenolics with a selectivity of 91% (birch wood, 250 °C, methanol).[24,96] Similarly, the solvent system impacts both the yield and composition of the resulting monomers. Song *et al.*[97] reported a 50 wt% yield of phenolic monomers in methanol, while lower yields were observed in ethanol (48 wt%) and isopropanol (27 wt%) (birch wood, 200 °C, Ni/C). More recently, Ouyang *et al.*[98] achieved a 49% yield of phenolic monomers with 82% selectivity for 4-propyl syringol using a methanol/water (1 : 2 v/v) mixture under hydrogen-free conditions (birch wood, 230 °C, Pt/Al_2O_3, methanol/water (1 : 2 v/v)). They also demonstrated that adjusting the MeOH/H_2O ratio and reaction temperature could shift selectivity towards propenyl or propyl-substituted monomers. Furthermore, the type of biomass used also has a strong influence on the product composition after depolymerization related to the syringyl/guaiacyl ratio and the content of extractives, acetates, or free sugars of the feedstock.[29]

As an alternative, defunctionalization strategies are gaining interest to improve the selectivity and purity of monomers from lignin depolymerization, providing a more sustainable approach to bulk platform chemicals. Typically, defunctionalizing the phenolic core is achieved through hydrodeoxygenation (HDO), yielding simpler products. The level of deoxygenation and defunctionalization depends on the reaction conditions, resulting in various end products, such as (*n*-alkyl) phenols, (*n*-alkyl)cyclohexanols, aromatics, or aliphatics.[30]

6.1.1. Phenol. Phenol, a crucial platform chemical used primarily for bisphenol A (BPA), phenolic resins, and polyamides, is typically produced from fossil-derived benzene but can also be synthesized from lignin-derived monomers (Fig. 7).[99] For example, the Sels group developed a two-step process involving demethoxylation with Ni/Si and dealkylation with zeolite catalysts, converting crude lignin monomers to bio-phenol and bio-propylene in yields of 20 wt% and 9 wt%, respectively, based on birch wood lignin.[92,100] Another approach by the Hensen group uses metal phosphides to convert 4-*n*-propylguaiacol from pinewood to phenols through demethoxylation and transalkylation using H-ZSM-5, yielding 9.6 mol% phenol from lignin.[101] Zhang *et al.* reported a similar conversion using a one-pot process with Pt on carbon and H-ZSM-5, achieving 60 mol% phenol from pure 4-*n*-propylguaiacol.[102]

Being a petrochemical, its prices fluctuate based on crude oil prices but have been in the range of €700–€1100 per ton in recent years.[103–105] Based on the lignin oil prices mentioned in section 4 (>€1600 per ton) and the above-given yields, it is clear that bio-phenol made from lignin oil-derived monomers will be several times more expensive than fossil phenol as a sole product, mandating the effective valorization of other product streams if economic parity is to be approached.

6.1.2. Catechol. Catechol is a valuable chemical, with applications in agrochemicals, polymers, flavors, fragrances, pharmaceuticals, and corrosion inhibitors.[106] Its industrial production from fossil benzene-derived phenol often suffers from low selectivity and harsh conditions.[107] However, lignin-derived monomers with two oxygen atoms on the phenolic ring offer a promising pathway to catechol (Fig. 7). For example, catechol can be synthesized through (1) *O*-demethylation of 4-*n*-propylguaiacol (4-PG) with acidic zeolites to form 4-*n*-propylcatechol, followed by (2) *C*-dealkylation to yield catechol and propylene. This approach yields approximately 70% catechol from pure 4-PG or 56% from crude 4-PG monomers from softwood RCF oil.[108]

As catechol is largely derived from fossil benzene, the market price of catechol also naturally fluctuates in sync with benzene prices, with a reported range between approximately €2000 and €4000 per ton.[105,109] The price of lignin-derived bio-catechol is likely to be higher than fossil catechol prices, although the difference might be less than in the case of phenol if the yields mentioned for the above route can be realized industrially.

6.1.3. Terephthalic acid. Terephthalic acid, essential for producing poly(ethylene terephthalate), is synthesized from depolymerized RCF lignin oil through a multi-step process: (i) demethoxylation with Mo on carbon, (ii) carbonylation of *n*-alkylphenols with CO using a Pd catalyst, and (iii) oxidation to terephthalic acid with a Co–Mn–Br catalyst under oxygen pressure. This process, starting from lignin oil derived from corn stover, achieved a 65.7 mol% yield of 4-*n*-alkylphenol post-demethoxylation. Subsequently, carbonylation and oxidation yielded 4-alkylbenzoic acid and terephthalic acid at 75 mol% and 60 mol% respectively, with a final terephthalic acid yield of 30 mol% (15.5 wt% based on corn stover lignin).[110]

The yields that are currently achieved in the conversion of lignin oil to terephthalic acid are not expected to be cost competitive with fossil terephthalic acid for which €1000–€1400 per ton is a typical price range for purified terephthalic acid[111,112] – and further optimization is required.

6.2. Biofuel

The aromatic chemical structure of lignin combined with its large-scale availability make it a potential source for sustainable biofuels (Fig. 7). The current market for biofuels is dominated by bioethanol, produced from the conversion of corn, and biodiesel, from the hydrogenation of palm and used vegetable oil. Lignin-derived biofuels can offer a lower carbon footprint alternate to bioethanol, helping decarbonize hard to electrify sectors such as sustainable aviation fuels.[113,114]

The products of traditional hydrogenolysis are mixtures of phenolics (*i.e.*, cresols, guaiacols, syringols, catechols, *etc.*) and thus not suitable for direct use as drop-in biofuels. Thus, a subsequent catalytic hydrotreatment step is necessary to upgrade the hydrogenolysis oil. The catalytic hydrotreatment of lignin oils has been extensively studied experimentally using a range of catalysts such as supported noble metal catalysts (Pd, Rh, Ru, Pt),[115–117] commercially used hydrodesulphurization (HDS) catalysts such as $NiMo/Al_2O_3$ or $CoMo/Al_2O_3$ [118–121] and relatively inexpensive supported Ni,[117] Fe[122] and Mo_2C catalysts.[123]

In addition to using hydrogenolysis oils, alternatives to produce biofuels from extracted lignin also exist. A recent example shows the use in a one pot catalytic hydrocracking approach[73] in which the harsher depolymerization temperature combined with the use of zeolites help cleave C–C linkages in the lignin molecule to enhance the monomer yield. The produced bio-oils had higher monomer yields compared to RCD (up to 54%) significantly increasing its potential for use as biofuel. More recently, bifunctional modifications to zeolites, incorporating both a Lewis acid and hydrogenation sites using Ni_1Zn_1/HY_a and Co–Zn/off-Al H-beta was reported to depolymerize lignin into liquids with high yields and low char formation.[124]

The large variety of fuels in commercial use provides numerous potential entry points for lignin-derived biofuels but also makes it hard to provide a single target benchmark price. Depending on the fuel grade and market conditions, the prices of automotive fuels like diesel and gasoline tend to be around €700–€800 per ton,[125] marine bunker fuel prices are typically lower (€600–€750 per ton),[126] and jet fuel prices roughly similar to automotive fuels (€700–€850 per ton).[127,128] The most-widely produced biofuel (*i.e.*, bioethanol) costs €400–€700 per ton for first-generation feedstocks and €800–€1200 per ton for advanced (lignocellulosic) feedstocks[129] and is already used as a renewable replacement in gasoline. Aviation biofuels, however, are harder to synthesize economically, as they consist of longer carbon chains, and consequently cost almost €3000 per ton.[127,128] For this reason, lignin-based sustainable aviation fuels, with an estimated production cost around €1200–€2400 per ton have been gaining interest.[130,131] However, optimization is required to improve the process efficiency.

6.3. Polymer applications and additives

As lignin is characterized by an abundance of aliphatic and aromatic hydroxyl groups, it is a highly interesting renewable resource for a wide range of material applications (Fig. 7). Lignin can potentially improve mechanical and rheological properties of polymer networks and blends,[132] but the unique molecular structure of lignin also adds antioxidative, antimicrobial and antistatic properties as well as improved UV stability to materials.[133,134] However, replacing fossil polymer building blocks and additives with lignin requires ideally a feedstock of lower polydispersity and molecular mass (compared to technical lignin side streams) as well as control over functional groups to overcome issues related to a very heterogenous molecular structure.[135] Nevertheless, it is not necessary to fully convert lignin into monomers for this purpose. It has been demonstrated that oligomeric fractions of depolymerized lignin are good enough or sometimes even better suited to obtain plastics or resins of satisfactory properties.[136] The focus of the following short section will be exclusively on how the molecular structure of lignin and lignin oils prior to subsequent modification or derivatization steps affects their use as polymer building blocks and additives in polymer matrix materials. Several more detailed and comprehensive reviews of the prospects of lignin for material applications have been published throughout the last years, covering aspects beyond the scope of this work.[132,137–142]

The main problems associated with directly utilizing polymeric technical lignins with complex and heterogenous molecular structures[143–145] for the synthesis of, *e.g.*, biobased epoxy resins are the limited solubility in organic solvents and the suboptimal reactivity upon glycidylation.[95,146] Due to the structural similarity of lignins with epoxy resin networks, numerous studies have been conducted to replace the hazardous BPA of petrochemical origin with renewable and safer lignin-based alternatives.[142,147] Moreover, the estimated minimal selling price of the lignin bio-oil would be close to the BPA price range (*i.e.*, between €1100 and €1700 per ton in 2024).[148] Besides solely enhancing the material value by increasing the biobased carbon content of epoxy resins, introducing a glycidylated well-defined, low molecular mass fraction of lignin into the resin formulation improves the mechanical performance compared to a reference based only on BPA.[149] It has been clearly demonstrated that lignin of lower molecular mass and a high content of aromatic hydroxyl groups is beneficial to obtain epoxy resins featuring a higher Young's modulus (*i.e.*, a more rigid material).[150] Additionally, the mechanical properties of lignin-based epoxy resins can be further enhanced by a smart selection of the hardener.[151]

Besides introducing lignin into epoxy resins, the introduction of lignin into the network of phenol formaldehyde (PF) resins has been extensively studied to obtain, for example, wood adhesives with a higher renewable carbon content.[152–154] Same as for epoxy resins, a resin of better mechanical properties has been obtained by incorporating lower molecular mass fractions of lignin into a PF adhesive.[155] However, it seems that this trend does not extend all the way to the utilization of lignin oils predominantly composed of monomers and dimers. In another study, the incorporation of an oligomeric fraction of depolymerized lignin into PF resins resulted in adhesives exhibiting better mechanical properties than the incorporation of the lowest molecular mass fraction.[156] At the same time, both materials prepared with depolymerized lignin fractions performed better than the material prepared with polymeric lignin. However, as explained in section 6.1, a phenol substitute obtained from depolymerized lignin cannot currently be offered at a competitive price given the phenol prices of recent years (700–1100 € per ton).[103–105]

Lignin has also received attention as a renewable polyol replacement in polyurethane (PU) applications in which the minimal selling price of the bio-oil is estimated close to fossil-based alternatives (1200–2000 € per ton).[157] Within the available studies, the impact of the molecular structure of lignin on the preparation of PU coatings has been studied as well through combining solvent fractionation and depolymerization.[74] The conclusion was that high molecular weight lignin fractions deliver a PU coating characterized by a high glass-transition temperature (T_g), rigidity, and hydrophobicity whereas lower molecular mass fractions afford more hydrophilic and flexible coatings with a low T_g. The coating prepared from depolymerized lignin had similar properties as the coatings prepared with low molecular mass lignins obtained through solvent fractionation. However, due to a high content of aliphatic hydroxy groups formed upon depolymerization and the lower polydispersity, an increased reactivity of the lignin fraction with the isocyanate linker was observed. Besides that, the nature of the isocyanate linker also influences the reactivity of certain lignin hydroxy groups.[158]

As an alternative to incorporating lignin covalently bound into a thermoset polymer network, lignin can also be used as additive and functional filler material. In many studies, lignin particles were blended with thermoplastics to improve their mechanical and thermal stability.[132,141] In terms of the mechanical performance, the particle size of lignin is more important than the molecular weight, as the particle size strongly influences the dispersion of the lignin in the matrix material. However, a lower molecular mass has been related to better antioxidant properties of lignin/polymer blends.[159] This observation seems to be a direct consequence of a molecular structure characterized by an elevated phenolic hydroxy group content.[133] The functional groups available in the molecular structure of lignin also determine the interfacial adhesion between lignin and matrix material which in turn defines the thermal and mechanical properties of the blend.[160] For a polar matrix polymer an abundance of hydroxy groups is certainly beneficial but nonpolar matrix polymers, like polyolefins, require lignin particles characterized by a different surface chemistry.

6.4. Global market context

Thanks to the ubiquity of fossil-derived chemicals and fuels, biomass-based platform chemicals and fuels could have very wide applicability in theory. However, the total potential demand for biomass far exceeds its sustainable supply, necessitating a highly prioritized allocation. Based on the unavailability of alternatives and cost-effectiveness of bio-based solutions, previous work has identified materials production and aviation biofuels as the highest priority uses for a sustainable biomass supply.[161] An illustrative scenario aiming to stay within sustainability limits suggests that meeting the demand for these prioritized sectors would require approximately 54 EJ per year of biomass by 2050, a figure within the estimated prudent sustainable supply range of 40–60 EJ per year. This implies that a substantial portion of the available sustainable biomass must be directed to these high-value applications to achieve meaningful fossil displacement.

Specifically for the chemical and derived materials sector, it has been estimated that biomass can sustainably provide 20% of the sector's overall carbon demand by 2050.[162] To meet this 20% share, the global biomass consumption for the chemical and derived materials industry is projected to be between 444 and 447 million tonnes dry matter by 2050, representing a quadrupling of the current biomass demand for this sector. This limited availability of biomass and abundance of potential applications makes its efficient and economical utilization all the more important. This extends to reductive depolymerization technologies, for which efficiently utilizing the phenolic products is essential for the economic viability and ultimate success of these methods.

6.5. Conclusion applications

As shown above, lignin bio-oils have a broad range of potential applications in the current industrial landscape. However, the cost increase linked to the use of lignin often requires additional advantages to facilitate the transition towards the bio-based alternative. While increased renewability and sustainability are present in platform chemicals and biofuels, a fast transition to their biobased variants is unlikely without external stimuli from policymakers. In this regard, the increased use of sustainable aviation fuels might require the necessary cyclic compounds generated by lignin and aid the implementation of this technology.[114] The bio-oil favored for these applications requires a high monomer content and low production costs. At this moment, RCF oil would be favored because of its high monomer content as RCD oils prepared from current commercial lignin (*i.e.*, technical lignin) will have a low monomer content. The higher monomer content will reduce the downstream processing required to transform the lignin to the desired platform chemicals or biofuels.

The use of bio-oils in polymer applications and additives, on the other hand, has additional advantages to sustainability such as improved material properties and reduced hazardousness. These advantages justify a price increase in comparison to fossil-based alternatives and the use of bio-oils in polymers can thus be expected to have a faster transition to industrial scale. In polymer applications, the best mechanical performance is often obtained with lower molecular weight oligomers, while a high monomer content could reduce the performance. In this regard, RCD would be a suitable technology as the presence of monomers in the bio-oil is lower and mainly shorter oligomers are obtained.

7. Conclusion

Replacement of fossil resources by renewable sustainable biomass carbon resources is essential to achieve a sustainable and controllable carbon cycle. In this aspect, the valorization of the lignin component within lignocellulosic biomass could be key for the replacement of fossil fuel-based aromatic platform chemicals. The two main reductive depolymerization techniques, RCF and RCD, which are currently considered for upscaling to industrial scale have been thoroughly discussed and compared within this review. From a technical side, RCF has the advantage of being a one-step process which decreases the energy and solvent consumption required within the process. On the other hand, RCD using an isolated lignin can be performed in a continuous way which is better suited for industrial valorization. Additionally, it reduces catalyst fouling by avoiding the interaction of the catalyst with the biomass pulp which should allow longer production times than for RCF.

From an economic perspective, the current models estimate similar to slightly lower prices for RCF than for RCD but further research will be key to decrease the minimum selling price. In this regard, RCF focused already on product recycling which significantly lowered solvent, CAPEX and steam costs of RCF. RCD, on the other hand, focuses its process improvements on increasing the lignin solubility as it improves the feedstock, solvent and CAPEX costs of the process. The feasibility of these improvements has to be proven scientifically, and strong efforts are needed to decrease the price even further to ease the transition from fossil-based feedstocks to bio-based feedstocks. Finally, from the product side, a significantly higher monomer content is currently present in the product stream of RCF (~25–45 wt%) in comparison to RCD (<10 wt%). However, RCD of lignin with high ether bond content (*e.g.*, AAF lignin) holds the potential to achieve similar monomer contents to RCF values. This would enhance the product flexibility of RCD, as both low and high monomer content RCD oils can be produced and thus increase the range of targeted applications.

The current difference in monomer content changes the applications which should be primarily targeted for the two streams. Some applications, such as platform chemicals and biofuels, favor bio-oils with a high monomer content, which are more suitable for the current RCF process. On the other hand, the presence of small oligomers can be beneficial for applications, such as plastics and additives, as it improves the mechanical properties in comparison to a pure monomer stream. For these types of applications, producing a stream with a lower monomer content and higher oligomer content *via* RCD would be beneficial. The difference between the target applications of RCF and RCD shows the complementarity of both approaches towards bio-aromatic production rather than their competitiveness. While clear market potential is available for both streams, further efforts are required to obtain processes that allow industrial scale production of sustainable aromatics and can be competitive to traditional fossil fuel-based processes.

Author contributions

BD: Conceptualization, writing – original draft, writing – review & editing. BS: Conceptualization, writing – original draft, writing – review & editing. PJ: Conceptualization, writing – original draft, writing – review & editing. AM: Writing – original draft, writing – review & editing. JC: Writing – original draft, writing – review & editing. JKK: Writing – original draft, writing – review & editing. MVD: Writing – review & editing. KV: Funding acquisition, writing – review & editing. PJD: Funding acquisition, writing – review & editing. MLS: Funding acquisition, writing – review & editing. EF: Conceptualization, Funding acquisition, writing – review & editing.

Conflicts of 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.

Data availability

The data supporting the techno-economic analysis in this article have been included as part of the SI. Supplementary information is available. See DOI: **https://doi.org/10.1039/d5gc02316e**.

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