

Review

Recent Advances in Lignin-Based Biofuel Production

Engin Kocaturk ^{1,2,*}, Tufan Salan ^{2,3} , Orhan Ozcelik ⁴, Mehmet Hakkı Alma ⁵ and Zeki Candan ^{2,6} ¹ Department of Nanotechnology Engineering, Zonguldak Bulent Ecevit University, 67100 Zonguldak, Turkey² Biomaterials and Nanotechnology Research Group & NanoTeam, 34473 Istanbul, Turkey³ Department of Materials Science and Engineering, Kahramanmaraş Sutcu Imam University, 46050 Kahramanmaraş, Turkey⁴ Department of Aerospace Engineering, Ankara Yıldırım Beyazıt University, 06010 Ankara, Turkey⁵ Department of Biosystem Engineering, Iğdir University, 76000 Iğdir, Turkey⁶ Department of Forest Industrial Engineering, Istanbul University Cerrahpasa, 34098 Istanbul, Turkey

* Correspondence: enginkocaturkk96@gmail.com; Tel.: +90-372-6436601; Fax: +90-372-6436604

Abstract: Lignin is a polymer found in the cell walls of plants and is an important component of wood. Lignin-derived fuels have attracted attention as a means of producing biofuels from biomass in recent years. There are two basic methods for converting lignin into fuel: thermochemical and catalytic. Lignin-derived fuels have the potential to reduce dependency on fossil fuels and reduce greenhouse gas emissions. However, more research is needed to optimize the production of lignin-derived fuels and to determine their environmental impact. This review aims to evaluate the development of lignin-derived fuels from an economic and environmental point of view while presenting a broad perspective.

Keywords: lignocellulosic biomass; lignin-derived fuels; biofuels; lignin engineering



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1. Introduction

Lignocellulosic biomass is a promising source of raw materials used to produce biofuels. The rapid depletion of fossil fuel resources in the world and the increase in the environmental effects of these resources have led to the use of sustainable energy resources instead of fossil fuels. The formation of toxic gases by fossil fuels pollutes the environment and causes global warming [1–5]. The use of lignocellulosic biomass in biofuel production not only reduces emissions, but also helps to reduce greenhouse gases. In addition, the renewability and abundance of lignocellulosic biomass is very important for environmentally friendly biofuel production [6–12]. Today, biomass-based fuels are commercially available. For example; The number of bioethanol (made from cellulose and hemicellulose-based carbohydrates) and biodiesel (produced using vegetable oils) fuel stations is increasing in the USA and Canada. However, considering price–performance comparisons and purity levels, the quality of the resulting fuel needs to be improved. In the literature, biomass-based fuels are evaluated under two basic classes as furanic biofuels and cycloalkane biofuels. Furanics use the dehydration of carbohydrates to produce compounds with a furanic structure. Cycloalkanes are produced by hydro-deoxygenation of lignin with the help of lignin phenolic compounds. Cycloalkanes obtained using lignin can be used directly as fuel and mixed with conventional gasoline. Thus, a reduction in fuel density, volumetric heating, and NO_x emissions can be achieved by using conventional gasoline–cycloalkane biofuel mixtures [13–20]. Lignin-derived biofuels have the potential to be a sustainable alternative to fossil fuels, as they can be produced from renewable plant materials and have a lower carbon footprint. Overall, the potential for lignin-derived biofuels is promising, but more research and development are needed before it becomes a practical and economically viable alternative to fossil fuels. Lignin can be converted into a range of fuels, including gasoline, diesel, and aviation fuel. These fuels have similar properties to their fossil-based counterparts, making them suitable for use in a wide range

of applications. In addition, lignin-based fuels can be blended with traditional fossil fuels, helping to reduce the overall carbon intensity of the transportation sector [21–23]. Conventional aviation fuel is derived from fossil fuels, which contribute significantly to greenhouse gas emissions. Lignin-based aviation fuel offers a renewable alternative that can reduce emissions and improve air quality. Lignin-derived aviation fuels have the potential to significantly reduce our dependence on fossil fuels and reduce greenhouse gas emissions. Although still in the early stages of development, lignin aviation fuels have shown positive results in laboratory tests and have great implications for the future of aviation [24–26]. Biofuels obtained by using lignin can be used in many different fields (heat, electricity, and fuel production) [27–33]. Today, the future of lignin-based biofuels is possible by understanding the basic elements and mechanism of lignin-based biofuels [34]. In this context, this review article will examine the economics and sustainability of lignin biofuels, as well as the processes and mechanisms necessary to understand the role of lignin in biofuel production, and contribute to the industrial design of lignin-based biofuels in the future.

2. Lignin and Its Derivatives

Lignocellulosic micro and nano materials derived from natural lignocellulosic biomass contain three basic components. They consist approximately of 35–50% cellulose, 5–30% lignin and 20–35% hemicellulose by weight [35,36]. Lignin is known as the second most abundant naturally occurring organic substance on Earth after cellulose [37,38]. It disperses around the cellulose fibers in the plant cell wall together with the hemicellulose. In addition, lignin is covalently bonded to the carbohydrate/cellulose structure [39–42]. It is known that the total amount of biomass in the world corresponds to an estimated $1.85\text{--}2.4 \times 10^{12}$ tons, and approximately 20% of the total biomass amount is lignin [43]. Although it varies according to the extraction process, it has a molecular mass of $1000\text{--}20,000 \text{ g mol}^{-1}$ [44,45]. Lignin and lignin derivatives can be used to obtain many products with high added value, especially carbon fiber, some oxidized products, phenolic compounds, synthesis gas, various hydrocarbons, and biofuels [46–48]. There are numerous properties which make lignin qualify as a unique natural resource. These properties include biodegradability, structural integrity, antioxidant/fungal/microbial behavior, abundance, chemical/biological/enzymatic stability, fire/UV resistance, and hydrophilicity/hydrophobicity (depending on the lignin source). Moreover, lignin is known to possess excellent physicochemical, rheological, and viscoelastic properties, strong film-forming capacity, and good compatibility with many industrial chemicals compared to cellulose and hemicellulose [49–55]. Despite the above-mentioned outstanding properties, utilization of lignin at an industrial scale is still difficult, primarily because of its impurity, chemical reactivity, and delicate structure [56]. Depending mainly on the isolation method, lignin is classified into several species and derivatives. Figure 1 presents a summary of lignin species under four main headings. Lignin derivatives and isolation mechanisms typically used, along with the most general approach, are kraft lignin [57], lignosulfonate lignin [58], soda lignin [59], organosolv lignin [60], isolated lignin [61], acid hydrolysis lignin [62], ball-milled lignin [63], and degraded lignin [64].

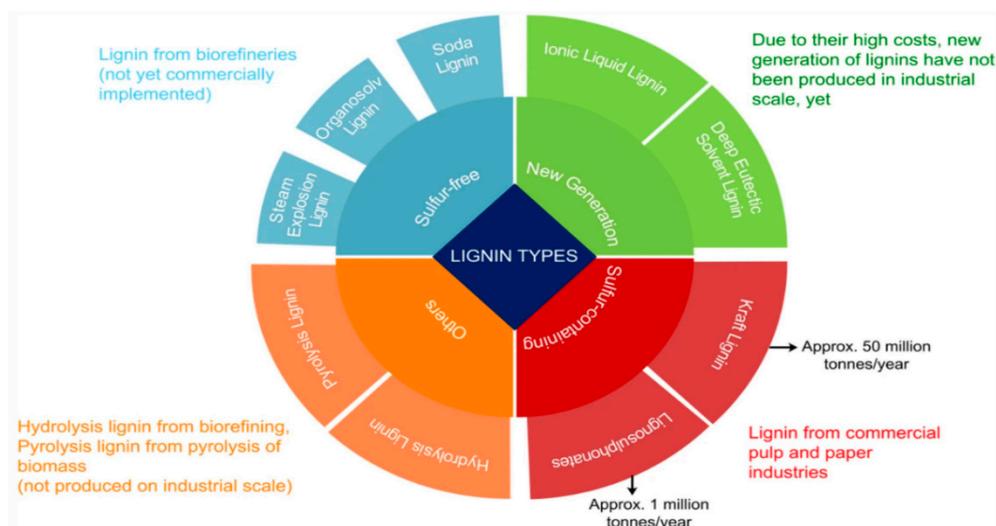


Figure 1. The most common lignin derivatives known in the literature, their sources, production, and classification [65].

3. Modification and Pretreatment of Lignin for Improved Biofuel Conversion

Lignocellulosic biomass has the potential to be a valuable resource for the production of biofuels, but its conversion into useful products is hindered by the presence of lignin. Lignin is a complex polymer that is difficult to degrade and therefore generally needs to be modified and pretreated before it can be used for biofuel production. The process of modifying and pretreating lignin is important for improving the efficiency of biofuel conversion from lignocellulosic biomass. Pretreatment can improve the digestibility of lignocellulosic biomass by breaking down the complex polymeric structure of lignin, thus enabling the release of sugars and other valuable compounds [66,67]. Acid, alkaline, and enzymatic hydrolysis techniques are used for pretreating lignin. Acid and alkaline hydrolysis involve the use of acids or bases to break down the polymer structure of lignin, while enzymatic hydrolysis involves the use of enzymes to catalyze the degradation of lignin [68,69]. Pretreatment is an important step in the process of converting lignocellulosic biomass into useful products, and it can have a significant impact on the economics of biofuel production and is essential for improving the efficiency of biofuel conversion from lignocellulosic biomass. The cost of pretreatment is directly related to the price of lignocellulosic biomass, as the cost of pretreatment is often proportional to the amount of lignin that needs to be broken down [70,71]. Additionally, pretreatment can also reduce the energy requirements for biofuel conversion, as breaking down the lignin requires less energy than that of cellulose and hemicellulose. In order to maximize the potential of lignocellulosic biomass for biofuel production, it is important to understand the different strategies for pretreating lignin, and their impact on the biofuel conversion of lignocellulosic biomass. By exploring the different strategies for modifying and pretreating lignin, it is possible to reveal the potential of lignocellulosic biomass for biofuel conversion [72,73].

Different Pretreatment Strategies of Lignin

Acid hydrolysis is one of the most common methods of extracting lignin from lignocellulosic biomass. Acid hydrolysis involves the use of acids such as sulfuric acid, hydrochloric acid, and nitric acid. This method involves the use of acid to break down the bonds of the lignin molecule. The relative amount of lignin increases with longer reaction times and/or greater acid concentrations for all biomasses. The acid hydrolysis process is used to produce sugars and biobased platform chemicals from lignocellulosic biomass. Additionally, acid hydrolysis is used to produce lignin-derived solid acids (LDSAs) and to extract furfural and acetic acid from hot water hydrolysis liquor. A comparative study of acid hydrolysis of lignin and polysaccharides in biomasses was conducted to investigate

the effects of different acid hydrolysis conditions. The results showed that lignin reactions (dissolution/deposition) occurred in a greater extent for bagasse and straw than for eucalyptus, considering similar conditions of acid hydrolysis. The sugar content quantified in eucalyptus decreased as the acid concentration and/or reaction time in the second hydrolysis increased. It was also found that glucose, galactose, and mannose were more resistant to harsher acidic conditions than xylose and arabinose [74].

The alkaline pretreatment is another chemical pretreatment technique which uses chemicals such as ammonia (aqueous, liquid, and gaseous), sodium hydroxide, sodium carbonate, and calcium hydroxide (lime). Alkaline reagents primarily interact with lignin; as a result, they are more effective at removing lignin. Pretreatment chemicals that are alkaline are less caustic than those that are acidic, like sulfuric acid and sulfite. As demonstrated by soaking in sodium hydroxide or ammonium hydroxide, alkaline pretreatments are performed under milder conditions, some of them even at ambient temperature. By using these techniques, the need for expensive materials and specialized designs to combat corrosion and harsh reaction conditions may be avoided [75].

Enzymatic hydrolysis is yet another strategy for pretreating lignin. This process involves the use of enzymes like ligninases, cellulases, and hemicellulases to catalyze the degradation of lignin. Enzymatic hydrolysis is often used in conjunction with other pretreatment strategies, such as acid and alkaline hydrolysis. Enzymes are an important component of lignocellulosic pretreatment, as they are used to catalyze the breakdown of lignin. Enzymes, however, can be expensive and have a limited lifespan, as they are often denatured by the pretreatment process. As such, it is important to examine the reusability of enzymes in order to maximize the efficiency of lignocellulosic pretreatment [76,77].

Physical methods, such as mechanical grinding, mechanical extrusion, microwave, and pyrolysis can also be used to modify lignin. These methods can be effective in breaking down lignin into smaller pieces, but they are also energy-intensive and can generate unwanted by-products [78]. In addition to these methods, there are also other strategies for pretreating lignin, such as biomass sonification. Biomass sonification involves the use of ultrasound to break down the complex polymer structure of lignin, thus resulting in the release of valuable compounds and improving the digestibility of lignocellulosic biomass [79]. Biomass sonification is an emerging technology and has several benefits, including lower energy requirements, higher sugar yields, and lower costs. The lower energy requirements of biomass sonification make it an attractive option for the pretreatment of lignin. In addition, biomass sonification can also increase sugar yield from lignocellulosic biomass, thereby providing higher biofuel yields, making it an attractive option for biofuel production [80,81].

Biological methods, such as the use of microorganisms, can also be used to modify lignin. Microorganisms like fungus can break down lignin through a process called biotreatment, which involves the secretion of enzymes that digest lignin. This method is environmentally friendly and can be more cost-effective than chemical or physical methods. However, the biotreatment method is limited by the low biofuels yield [82].

4. Lignin-Based Biofuels via Thermochemical Routes

The conversion of lignin to high value-added chemicals and materials via thermochemical methods has been an attractive research theme in the literature, because these methods are convenient for industrial scale production with relative ease and more economy [83–85]. Thermochemical conversion techniques of lignocellulosic biomass include various conversion techniques such as pyrolysis, hydrothermal carbonization, and gasification, and Table 1 shows the minimum process requirements and post-processing products required to perform these techniques [86–91]. These thermochemical techniques are advantageous over the direct combustion process in that they release a lower amount of hazardous gases and enable the formation of high value-added compounds during the conversion process [92,93].

Table 1. Thermochemical pathways used to convert biomass to biofuel, process requirements, and post-process products.

Process	Process Requirements	Product	Advantages and Disadvantages
Hydrothermal Carbonization	180–250 °C several hours	Gas Liquid Hydrochar	<ul style="list-style-type: none"> • It does not require pretreatment or much energy compared to pyrolysis, and the moisture content can be applied directly to the lignin. • It is possible to produce multifunctional materials with chemicals with high added value. • It requires high pressure for the reactor. • Investment costs are high.
Pyrolysis	350–700 °C >30 min	Bio-oil Biochar Gas	<ul style="list-style-type: none"> • It requires lower operating costs compared to other thermochemical processes. • Generally, it is possible to obtain high oil yields of approximately 75–80% after the pyrolysis process.
Biomass Liquefaction	300–400 °C 0.2–1.0 h 5–20 MPa	Bio-oil	<ul style="list-style-type: none"> • It can be applied to many raw materials. • The biomass used does not need to be dried. • The reactions take place at low temperatures. • It has no commercial use. • Dilute concentration is required for the process.
Supercritical Fluid Extraction	250–400 °C	Bio-oil	<ul style="list-style-type: none"> • The process has the convenience of working continuously. • Biomass has a higher conversion rate. • No catalyst is needed during the process. • Operating costs are high.
Gasification	>700 °C atm pressure	Syngas Fuel gas	<ul style="list-style-type: none"> • In energy recovery, it is possible to produce products with high efficiency, high added value, and generally less emissions; complex technology. • It requires high investment, production, and operating costs. • It is usually implemented on a pilot scale.

4.1. Hydrothermal Carbonization

The literature meaning of hydrothermal carbonization refers to the chemical and physical transformation of lignocellulosic biomaterial in environments containing high heat and pressure [94]. When lignin is subjected to hydrothermal treatment, its solubility increases. In addition, it interacts more quickly with water on a physical and chemical level and forms carbonaceous structures at the end of the process. The hydrothermal carbonization process includes demethoxylation, alkylation, condensation, hydrolysis, and cleavage of carbon–oxygen–carbon and carbon–carbon bonds [95,96]. While the bonds in the aromatic rings are not split in the hydrothermal carbonization process, the ether bonds and carbon–carbon bonds are cleaved in the hydrothermal carbonization process of lignin [97,98]. Compared to the pyrolysis process, hydrothermal carbonization can be directly applied to the high-moisture lignin without requiring pretreatment or much energy. However, hydrothermal carbonization technology is not widely used in industry. This is attributed to a number of problems encountered in the industry such as high pressures required in the reactor, high investment costs, and the wall effect in the heat–mass transfer during the process. Hydrothermal carbonization technology, although it has some negative aspects, can be applied for the production of value-added chemical materials and multifunctional materials obtained from the lignin and its derivatives [99–101]. Kim et al. [102] used the hydrothermal carbonization (HTC) method to obtain biofuel from lignin. It has been determined that the properties of hydrocoal produced with the help of HTC have characteristics similar to coal or other coal-based materials that have the potential to be used in place of coal. With the hydrothermal carbonization process, an increase in the carbon content of the biomass can be achieved. Xiao et al. [103] reported that the carbon content of the biomass, which was determined to be approximately 48%, increased to ap-

proximately 72% with the increase in carbon content after HTC treatment. In another study, lignin showed almost twice the char-forming properties of the HTC process compared to hemicellulose and cellulose [104].

4.2. Pyrolysis

The products obtained via pyrolysis in large quantities are known as bio-oil, combustible gas, and biochar. Several steps are required for lignin to be used in pyrolysis [94,105]. In the fast pyrolysis process, carbonization in the lignocellulosic mass can be minimized. In addition, liquid efficiency may be greater in the rapid pyrolysis process [106–108]. The pyrolysis of lignin typically takes place at temperatures between 400 and 750 °C. At the beginning of the pyrolysis process, the first bonds to break from the biomass are ether bonds with weak bond strengths. The efficiency of lignin pyrolysis increases when the temperature rises above 400 °C. At temperatures above 400 °C, however, the decomposition process becomes complex and tar formation is observed [109]. The pyrolysis process is generally evaluated under two headings as slow pyrolysis and fast pyrolysis. The primary basic product in the slow pyrolysis process is biomass char. In rapid pyrolysis, the main product is typically a liquid called pyrolysis oil or bio-oil. Rapid heating (typically > 100 °C/s) of lignocellulosic biomass is required to obtain high-yield bio-oil. It has been reported in the studies that the pyrolysis oil yield can be increased up to approximately 70% with the pyrolysis processes. During the pyrolysis process of lignin, thermally stable products such as CO, CO₂, H₂O, volatile liquids, phenolic compounds, gaseous hydrocarbons, coal, and coke are generally obtained [110–115].

4.3. Biomass Liquefaction

Liquefaction of biomass can be applied to many raw materials. However, biomass liquefaction is not used commercially [116]. Hydrothermal liquefaction (HTL) is the process of converting biomass (aqueous) into liquid fuel by processing it with temperature and pressure [117]. In the HTL, water acts as a solvent. This also eliminates the need for biomass drying. Moreover, it allows reactions to occur at low temperatures. Hydrothermal liquefaction is typically performed for 0.2 to 1 h at temperatures of 300–400 °C and pressures of 5–20 MPa [118,119]. The bio-oils obtained by the HTL process are in semi-liquid form, with a dark smoky odor [120–122]. There are many HTL studies in the literature using different lignin compositions at different temperatures and pressures. Abdelaziz et al. [123,124], in a study by the HTL method, reported that depolymerization of kraft lignin was achieved at a lignin content of approximately 5% by weight under 130 bar pressure at temperatures lower than 250 °C. HTL, performed in another study, showed the bio-oil with 10% lignin content to be obtained with the aid of a catalyst at 300 °C with approximately 50% yield [125]. In a similar study, it was reported to obtain bio-oil with an efficiency of up to 60% at 280 °C [126]. Although the hydrothermal liquefaction (HTL) technology allows the phenolic compounds in the lignin structure to be easily separated from the body in the aqueous medium, the technology has limitations such as the need for high temperatures, dilute concentrations, and the need for a second hydroprocessing step in biofuel production [127]. It is the process of thermal decomposition of biomass under supercritical conditions at high pressure and temperatures of 250–400 °C [128]. With the help of supercritical fluid extraction, organic compounds in the biomass can be effectively separated. The biofuels obtained in this way are affected by various factors such as the duration, temperature, and pressure of the separation process [129]. To obtain biofuel, ethanol and methanol were used many times as organic solvents in the supercritical liquid extraction process. Apart from these solvents, 2-propanol and 2-butanol are also used as solvents in biofuel production [130,131].

4.4. Gasification

Another way to obtain biofuel from lignin with the help of thermochemical pathways is gasification. This process is performed at very high temperatures such as 850 °C and above. Combustible gases and synthesis gases are produced by the gasification process.

The gases produced from here can be used to obtain products with high added value, such as fuel. Gasification of lignin from a lignocellulosic biomass is currently being implemented on a pilot scale due to high investment costs and uncertainties in the production costs [132].

Among the current topics of the recent past are the degradation of lignin with the help of thermochemical pathways and the conversion of lignin into valuable chemicals and fuels as a result of degradation. Thanks to the content of high aromatic parts in the molecular structure of lignin, thermochemical depolymerization of lignin can be achieved, and many products with high added-value phenolic part content can be produced at high productivity rates. Efficient use of lignin can reduce greenhouse gas emissions both directly (in this way, biofuels, and chemicals can directly produce) and indirectly (lignin products can increase the viability of biorefineries) [133–138].

5. Lignin-Based Biofuels via Catalytic Routes

Pyrolysis oil derived using lignin has some disadvantages. Some of these disadvantages can be listed as being thermally unstable, poor volatility, low calorific value, and high coking tendency [139]. Because of these shortcomings, the oil obtained from the pyrolysis of lignin cannot be used directly as bio-oil [140]. Therefore, the pyrolysis oil needs to be upgraded via catalytic methods. The catalytic methods used to upgrade the pyrolysis oil are hydrodeoxygenation, zeolite cracking, and hydrogenation [141]. The ambient conditions required for the catalytic oxidation of lignin and the comparison of the catalytic process types are given in Table 2.

Table 2. Comparison of the catalytic pathways used to convert biomass to biofuel.

Process	Process Requirements	Advantages and Disadvantages
Hydrodeoxygenation	120–400 °C 10–20 MPa hydrogen pressure.	<ul style="list-style-type: none"> • It is environmentally friendly. • It is possible to obtain bio-oil containing compounds with higher heating value.
Zeolite Cracking	350–500 °C atmospheric pressure	<ul style="list-style-type: none"> • Zeolites can show catalytic activity up to 50% of their volume. • The production of aromatic compounds by the process is done without H₂ requirements. • The resulting product has a low heating value compared to hydrodeoxygenated oils.
Hydrogenation	Exposure to hydrogen in a liquid phase at 100 °C to 500 °C with suitable catalyst.	<ul style="list-style-type: none"> • High efficiency is obtained. • The resulting fuel burns easily. • It can be used in various industrial applications.

5.1. Hydrodeoxygenation (HDO)

Hydrodeoxygenation (HDO) is a catalytic method used in biofuel production to remove oxygen in the molecular structure. In the literature, the hydrodeoxygenation process is also referred to as hydro-pyrolysis, hydrocracking, hydro-treatment and hydrogenolysis. In this process, high hydrogen pressure is mainly used to eliminate the presence of oxygen [142–146]. Typically, the environmental conditions required for an HDO reaction to occur include temperatures of 120–400 °C and hydrogen partial pressures of as high as 10–20 MPa [142,146,147]. If the HDO process is to be carried out using a heterogeneous catalyst, temperatures in the range of approximately 200–500 °C are used [147]. The type of catalyst utilized during the HDO process is important. The choice of catalyst varies depends on effectiveness of hydrodeoxygenation [142,146]. Many different catalyst types such as Ru, Pt, CoMoS, Mo₂N, P, Co, WP, Ni₂P, Ni–Cu, Ni–Co have been used in hydrodeoxygenation (HDO) reaction [148–154].

Jan et al. [155] aimed to obtain cycloalkanes by hydro-pyrolysis of lignin. For this purpose, they used HZSM-5 and Pd/HZSM-5 catalysts in their study. The results showed that the catalyst with the presence of Pd positively affected the hydrogenation reaction, catalyzed the reaction and greatly increased the yield of the cycloalkanes. In recent stud-

ies [156,157], a catalytic HDO reaction was used to produce biofuels from lignin. A bifunctional catalyst with bimetallic properties was used for the HDO reaction. The findings showed that the catalyst used had the highest hydrocarbon selectivity compared to the bimetallic catalysts used in other studies.

One study reported fuel production from cyclopentanol by evaluating lignin alkylation and hydrodeoxygenation together. In the study, beech powder was depolymerized and lignin oil resulting from depolymerization was used to obtain fuel. The characterization results revealed that the phenolic groups in the lignin oil content were converted into a fuel-usable precursor by approximately 80% as a result of HDO. Moreover, the resulting fuel mixture was reported to have a material density of 0.91 g/mL at 20 °C and a freezing point of less than −60 °C [158].

Zhong et al. [159] prepared Ru-based catalysts in different formulations to evaluate them as biofuels and examined the fuel properties of the catalysts by comparing their hydrodeoxygenation performance on lignin-based phenolic compounds. When the results were examined, it was reported that the Ru-based catalyst with TiO₂ nanosphere offered the most effective hydrodeoxygenation performance (approximately 50% increase in hydrocarbon content in lignin oil upgrading).

5.2. Zeolite Cracking

Crystalline solids with microporous structures are called zeolites. Zeolites are known as industrial catalysts and are frequently used in petroleum refining applications. Zeolite catalysts can be used to develop bio-oil. This process is generally carried out at temperatures of 350–500 °C and at atmospheric pressure [160,161]. It is a pressure-free reactor with the low amount of hydrogen required for zeolite cracking. In addition, the coking problem encountered in the HDO process is more effective in zeolite cracking [162–164].

5.3. Hydrogenation

The hydrogenation process of biomass has received increasing attention in recent years owing to its potential as a renewable fuel source. The hydrogenation process of biomass involves the conversion of carbon-containing molecules into hydrocarbons by reacting them with hydrogen in the presence of a catalyst. Biomass generally refers to any type of biological material that can be used as fuel or as a source for the production of energy. Unlike fossil fuels (e.g., coal and natural gas), biomass is renewable and can be replenished through natural processes. The hydrogenation of biomass generates usable fuels, such as ethanol and biodiesel, which can be burned to obtain heat and generate electricity [165]. The hydrogenation of biomass converts complex organic molecules to simpler ones with correspondingly lower molecular masses. The resulting low-molecular-mass fuel burns easily and, thus, is used in various industrial processes. The most common method of hydrogenating biomass involves exposing it to hydrogen in a liquid phase in the presence of a catalyst at temperatures ranging from 100 °C to 500 °C. In this process, the organic molecules are converted into short-chained hydrocarbons that can then be further processed to yield other useful products, such as jet fuels. Depending on the desired product and the properties of the raw material, different catalysts such as ruthenium, nickel, iron, cobalt, and molybdenum can be used in this process [166,167].

Hydrogenation of lignin is a chemical process that involves the addition of hydrogen atoms to lignin molecules in order to alter their chemical and physical properties. By modifying the chemical structure of lignin, it is possible to increase its solubility, reduce its viscosity, and improve its thermal stability, all of which are important properties for the production of biofuels and other bioproducts [168]. There are several methods for hydrogenating lignin, including catalytic hydrogenation, which involves the use of a metal catalyst to facilitate the addition of hydrogen atoms to lignin molecules, and non-catalytic hydrogenation, which relies on the use of high pressure and temperature to induce the addition of hydrogen atoms [169]. Catalyst samples used for catalytic hydrogenation of the lignin, the temperatures in which hydrogenation is performed, and the yields of the

lignin are summarized in Table 3. One major challenge in the hydrogenation of lignin is the diversity of its chemical structure, which can vary significantly depending on the plant source and the processing conditions. As a result, it is difficult to predict the outcome of the hydrogenation process and to optimize it for specific applications [170]. Despite these challenges, the hydrogenation of lignin has the potential to provide significant benefits in the production of biofuels and other bioproducts. By modifying the chemical and physical properties of lignin, it is possible to improve the efficiency and yield of biofuel production, reduce the environmental impact of biofuel production, and potentially create new markets for lignin-based products [171]. Overall, the hydrogenation of lignin is an exciting area of research with significant potential for the development of sustainable, low-carbon technologies and products. As research in this field continues to advance, it is likely that we will see further progress and innovations in the use of lignin in biofuel and other bioproduct production.

Table 3. Effects of temperature-catalyst changes on yield during the lignin hydrogenation process.

Catalyst	Temperature	Yield	Reference
Ni/C	200 °C	54%	[172]
Pd/C with CrCl ₃	280 °C	78.2–83.2%	[173]
Ru/C	200–250 °C	76.7%	[174]
HZSM-5 with NaOH	220 °C	61%	[175]
W ₂ C/AC	260 °C	12.7%	[176]
Porous Cu–Mg–Al oxide	140–220 °C	63%	[177]

6. Environmental and Cost Impact of Lignin Biofuels

The method used in the literature to identify opportunities to improve the environmental performance of a product throughout its life cycle (end of life) is called life cycle assessment (LCA) [178]. Techno-economic analysis (TEA) is carried out with the LCA and is generally defined as having similar functional units with the LCA outputs. All the costs required for a large-scale commercial biorefinery can be determined using techno-economic analysis. It is possible to evaluate the environmental and economic impact of lignin production thanks to data obtained from tools such as life cycle assessment (LCA) and techno-economic analysis (TEA). When the literature is examined, LCA studies on lignin can be evaluated under two headings. The first can be specified as life cycle assessment studies on lignin extraction (usually kraft extraction processes), and the second as LCA studies on lignin-based products [179,180].

Budsberg et al. [181] investigated the LCA evaluation of hydrocarbon biojet fuel obtained by bioconversion of poplar biomass. In the study, global warming potentials (GWP) and fossil fuel uses (FFU) for produced biojet and petroleum fuels were determined separately. The GWP and FFU values for petroleum fuel ranged from approximately 32 to 73 g MJ⁻¹ CO₂ and 0.71 to 1.0 MJ MJ⁻¹ for biojet fuel, while the GWP and FFU values were 93 MJ⁻¹ CO₂ and 1.2 MJ MJ⁻¹, respectively. From this point of view, it has been reported in the study that biojet fuels obtained using poplar biomass cause a decrease in global warming potential (GWP) and fossil fuel use (FFU) compared to fossil-based jet fuel, and it is promising for the development of biofuels that are needed by the aviation industry in the future.

In a study, the possibility of using pyrolytic lignin as a fuel was examined techno-economically, and for this, a model biorefinery was created using LCA. When the modeling results were evaluated, it was calculated that the required cost for the biorefinery plant setup was approximately \$277 million. In addition, the greenhouse gas (CO₂) emission values of the simulated biorefinery resulted in a reduction of approximately 60% compared to the petroleum-based refinery [182].

Obydenkova et al. [183] studied the environmental economics (production and transportation cost) of lignin-derived transportation fuels. In their study, the researchers dis-

cussed the suitability of fast pyrolysis of lignin in vehicles used in the marine and automotive industries in two parts, together with LCA and TEA analyses. In the first part, rapid pyrolysis of lignin was examined for TEA. In the second part, the greenhouse gas emissions of biofuels obtained by using lignin were evaluated by LCA. According to the TEA results, it was calculated that the minimum production costs of lignin biofuels in the marine and automotive fields ranged from 10 to 13.5 \$/GJ, while the transportation costs in the same areas ranged from 14.4 to 18.1 \$/GJ. When the LCA results were analyzed, it was determined that the greenhouse gas emissions of all fuels were not at levels compatible with the US renewable fuel standard (RFS). When all the results were evaluated, it was stated in the study that the economical use of lignin-derived transportation fuels was beneficial.

Shahbaz et al. [184] investigated the TEA of biomass components in the yield of slow-pyrolysis products of components in lignocellulosic biomass (cellulose, lignin, hemicellulose). According to the analysis, the cost of lignin pyrolysis was more economical (lignin (\$110/t), cellulose (\$285/t), and hemicellulose (\$296/t)). Also, when comparing pyrolysis CO₂ emission values, lignin was approximately 4 times lower than other biomass components. When all the obtained results were evaluated, it showed that lignin pyrolysis was more advantageous compared to cellulose and hemicellulose under the specified conditions.

Shen et al. [185] demonstrated a new catalytic way to produce hydrocarbons from waste lignin for use in jet fuels. In addition, TEA of the new catalytic pathway was performed. TEA results reported that the lowest selling price of lignin jet fuel obtained by catalytic raising of waste lignin was approximately \$6.35–1 (10% discounted value).

Kumaniaev et al. [186] converted birch bark into biofuel to be used as a fuel for road and aviation applications, then an environmental assessment was made. When the LCA results were analyzed, it was reported that the life cycle greenhouse gases (GHG) released during the process amounted to 1.7 kg of CO₂.

Moretti et al. [187] examined in depth the life cycle evaluations of lignin and lignin derivative products in the literature. According to the results of the examination, it was determined that the LCA studies on lignin and lignin derivatives were limited. Only a few studies have been reported to consider the lignin utilization stage and lignin end of life. Moreover, it has been determined that almost all of the LCAs prepared on lignin have been investigated for their effect on climate change. In addition, it has been observed in the studies that the LCA data are obtained mainly from laboratory and process simulations.

7. Conclusions and Future Prospects

Lignin, a complex polymer found in the cell walls of plants, has long been considered a waste product in the paper and pulp industry. However, recent research has shown that lignin can be converted into a range of valuable fuels and chemicals, opening up new opportunities for the bioenergy sector. One of the main advantages of lignin-derived fuels is their potential to reduce reliance on fossil fuels. Lignin is a renewable resource that can be sourced from a variety of plant materials, including wood chips, straw, and corn stalks. As such, lignin-based fuels have the potential to significantly reduce greenhouse gas emissions, helping the fight against climate change. There are several potential benefits to using lignin as biofuel feedstock. These benefits include abundance, renewability, and carbon neutrality.

- **Abundance:** Lignin is a waste product of the paper and pulp industry, and is produced in large quantities. This makes it a potentially cheap and readily available feedstock for biofuel production.
- **Renewability:** Lignin is a renewable resource, as it can be derived from plants that can be grown and harvested repeatedly.
- **Carbon neutrality:** Lignin is composed of carbon, hydrogen, and oxygen, and when it is burned as a biofuel, it releases the same amount of carbon dioxide as was absorbed by the plants during their growth. This means that the carbon emissions of lignin-derived biofuels are considered to be carbon neutral.

There are also a number of other potential applications for lignin-derived fuels, for example, biochar, resins, and plastics. Despite the many potential benefits of lignin-derived fuels, there are also some challenges to their widespread adoption. One of the main obstacles is the cost of production, which is currently higher than that of traditional fossil fuels. However, as production processes are refined and scaled up, it is likely that the cost of lignin-based fuels will become more competitive with fossil fuels. Modification of lignin is an important factor in improving the biofuel conversion of biomass. While there are several methods available, each has its own limitations and challenges. Another challenge is the need for more research and development to optimize the production of lignin-based fuels. LCA and TEA studies on lignin biofuels are a new and promising approach to meet these needs. While there are challenges to be overcome, the future looks bright for lignin-based fuels and for the bioenergy sector as a whole.

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