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Urea-Formaldehyde Resin Systems Modified with Partially Pyrolyzed and Delignified Lignin: A Review

Hande Yılmaz[a*], Doğu Ramazanoğlu[b], Halil İbrahim Uğraş [c], Ayhan Tozluoğlu[d]

[a]*İstanbul Gedik University, Faculty of Engineering, Department of Industrial Engineering* *hande.yilmaz@gedik.edu.tr (*Corresponding Author)*
[b]*İstanbul Gedik University, Faculty of Engineering, Department of Materials and Nanotechnology Engineering, Türkiye*
[c] *İstanbul Beykent University, Faculty of Engineering, Department of Chemical Engineering, Türkiye halilugras@gmail.com.tr*
[d] *Düzce University, Faculty of Forestry, Department of Forest Industry Engineering, Türkiye ayhantozluoglu@duzce.edu.tr*

Abstract

Urea-formaldehyde UF resins are typical adhesives employed in the production of wood-derived products. Application of such fossil fuel-based resins that emit formaldehyde emissions endangers the environmental and human health conditions profoundly. Bio-based lignin polymer can be utilized as a replacement additive in UF resins because it has inherent adhesive and water repellent nature. When lignin is added into the structure of the resin, the C-N bonds are reduced through the creation of new bonds within regions containing methylene bonds. Raw lignin cannot be directly added to UF resins; it must undergo pretreatment due to its poor binding capacity and amorphous nature. Therefore, partial pyrolysis of lignin in the low temperature zone under inert atmosphere was suggested in this study. This reaction creates a longer chain structure by partial C-O bond fragmentation; thus lignin is altered. This method is likely to reduce the formaldehyde emissions, but the same chain structure will increase the water repellency and reduce the moisture content of lignin. Literature research reveals that lignin additives have great effects on resin viscosity, free formaldehyde content, adhesion quality and panel performance.

Keywords: Lignin; UF Resin; Depolimerization; Partial Pyrolysis; Adhesives.

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1. INTRODUCTION

The negative effects of fossil fuels on sustainability, the environment, and human health have been increasingly recognized in scientific and industrial circles in recent years [1]; [2]. As a result, an increasing number of studies have focused on finding renewable, environmentally friendly, and safer alternatives to traditional raw materials [3]; [4]. In particular, the wood-based materials sector has increased its interest in bio-based resources due to their natural renewability and relatively low carbon footprint [1]; [5].

Ever since they were first introduced in 1884, urea-formaldehyde resins have been used extensively as adhesives in the wood sector [6]. Favorable properties of the resins—high reactivity, quick gelation, economy, nondiscoloration, and long-term durability make UF resins some of the most common formaldehyde-based adhesives in the world. Although used for ages, there has been growing controversy about their reported toxicological profiles, resultant implications, and recyclability-related issues. Huge efforts have been made in developing new formulations for UF resins that can reduce risks related to environmental sustainability and human health and support enhanced overall sustainability [7]; [8].

The high moisture resistance characteristic of urea-formaldehyde UF adhesives adversely impacts the performance and durability of wood products that rely on them, which makes optimization through formulation improvements essential [7].

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Towards this end, researchers have embarked on studies whose focus is reducing the formation of unwanted compounds, including toxic and non-recyclable high levels of formaldehyde, which are major human health hazards, while, at the same time, looking to enhance strength properties and improve the beneficial traits of adhesives [9]; [10]. Consequently, considerable focus has been on examining the use of bio-based lignocellulosic chemicals in UF resins. Lignin, the third most common lignocellulosic component, has attracted specific interest in these study efforts [10].

Due to the benefits of lignin, current research studies have sought to partially or entirely replace traditional raw material for adhesives such as phenol and melamine with lignin. However, the usage of lignin as an adhesive is regarded as more widespread and more extensive studies should be carried out in this area. Lignin's chemical composition is amorphous and complex. How pure lignin can be used and how it’s complicated structure can be retained has already been studied. In addition to these studies, it is also possible to reduce formaldehyde emissions by depolymerizing lignin into phenol units and using it as a monomer, and to increase its binding capacity with formaldehyde by converting it into a long chain [11]. However, it will also be expected that the water resistance of lignin will be improved due to the same chain structure [12]. Due to this reason, this study will attempt to review literature so that the role of lignin in UF resin can be discussed through the modification of the bond structure.

2. BASIC COMPONENTS OF PLANTS LIGNOCELLULOSIC STRUCTURES

Lignocellulosic structures consist of cellulose, hemicellulose, and lignin, the major plant cell wall and biomass components [13]. Cellulose is a glucose polymer consisting of long chains of glucose units linked to each other through β-1,4-glycosidic bonds. Hemicellulose is a heteropolymer consisting of the sugars xylose, mannose, galactose, and arabinose. Lignin is a phenylpropanoid polymer and an energy content enhancer compound for biomass [14]. These are the feedstock materials such as agricultural residues, forest residues, wood wastes, and energy crops that have immense potential for sustainable energy production along with the endless provision of bio-based chemicals and materials. A major benefit of lignocellulosic composition is that it does not compete for food resources and aligns with environmental sustainability as a carbon-neutral source; hence, such factors have placed lignocellulosic material top in the priority list within the bioeconomy [15].

Lignocellulosic biomass-based products not only have uses in the energy production but also in many others involving the composite materials, bioplastics, activated carbon preparations, adhesives, as well as resin processing [16]; [6]. Exploitation on the structural property auspitable in the aromatic compounds, specifically lignin, is predicted to prevail the next-generation material production development on the next-generation fiber reinforced biocomposites [17], among many other sustainable strategies. In addition, the studies on the structural change on the lignin-based products have also garnered significant interest, specifically on the consideration on the embracement on the advanced characterization techniques involving the scanning electron microscopy SEM, among others, with the aim to optimize the heat treatment conditions for the establishment on the quality on the resulting lignocellulosic biomass-based products [18]. During the previous decade, investment has been on the needs to reduce the reactors downsize, optimize the process for the defibrating, besides carrying the correct test on the products for the eventual exploitation on the lignocellulosic feedstock on the industrial scale [19]. Following the foregoing, the Plant Fundamental Constituent Ratios representing the proportional distribution between cellulose, the hemicellulose, as well as the lignin in the lignocellulosic biomass, are indicated in the following table.

Table 1. Plant Fundamental Constituent Ratios.

2	Component	3	Proportion on dry matter basis
4	Cellulose	5	20% - 50%
6	Hemicellulose	7	15% - 35%
8	Lignin	9	10% - 30%
10	Extractives	11	1% - 20%
12	Minerals	13	1% - 5%
14	Protein	15	5% - 15%
16	Fats	17	1% - 5%
18	Starch	19	5% - 30%

1.1 Lignin

The third most common component of lignocellulosic biomass is lignin. It is an amorphous polymer containing a phenylpropanoid unit [20]; [21]. The compound acts as a protective agent against microbial biomass degradation and increases the water resistance of plant cell walls. [22] & [23]. It is estimated that 225 million tonnes of industrial lignin are produced per year worldwide. [24]. Lignin is a by-product of biofuel production and is being used for the production of biomass energy [25]; [26].

The predominant content of plant cell walls is lignin. It is accompanied by cellulose and hemicellulose as the third most abundant wood-derived product biopolymer [27]. The biopolymer comprises phenylpropanoid units and has a complex and irregular structure [28]; [20]. Lignin has many notable characteristics such as high heat stability and strength due to its inherent polyphenolic structure [29]; [30]. The paper and pulp industry annually produces approximately 50 million tonnes of lignin [26]; [25].

This natural biopolymer occurring spontaneously in lignin holds significant potential for the reduction of the emission of formaldehyde [31]. With its complex aromatic structure containing units of phenylpropanoid, lignin possesses distinctive properties appropriate for both chemical as well as thermal conversions [28]. Because of this, it can simply be embedded in the matrices of polymers. In its resulting composite material, there is potential for the heat resistance to increase as well as for the gain in overall strength [30];[32]. Figure 1 below depicts the amorphous nature of the lignin, with an insistence on its disordered complex organization.

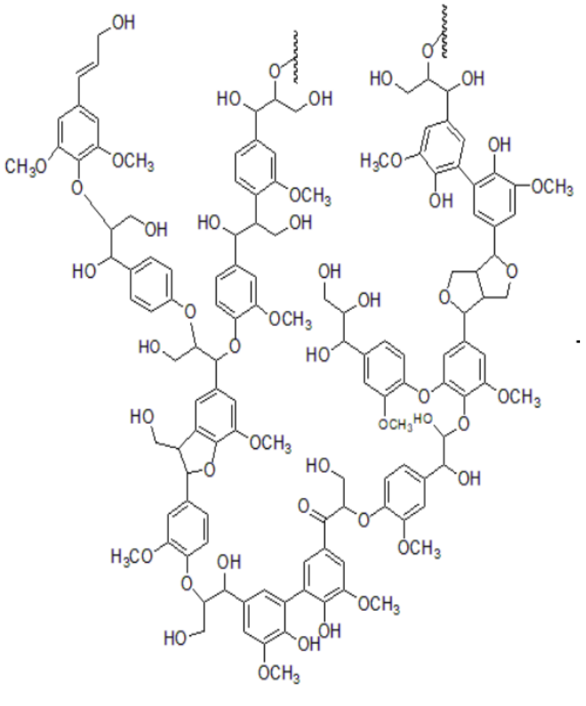

Figure 1. The amorphous structure of lignin.

3. DEPOLYMERIZATION PROCESSES

An essential step in the environmentally friendly synthesis of biofuels and biobased chemicals from plant biomass is the lignin depolymerization process [33]; [34]. In plant cell walls, native lignin is composed of phenylpropanoid units that combine to form an intricate polymer with mechanical strength and stiffness. However, its complex architecture poses limitations in biomass processing for biofuel generation [35]. Therefore, it becomes important to identify several methods that include thermochemical, catalyst, biological, and environmentally friendly solvent depolymerization methods, since depolymerization of the lignin through its breakdown into low molecular weight aromatics gives it greater value as a chemical feedstock [34]; [36].

3.1 Pyrolysis

Lignin pyrolysis is generally conducted in an oxygen-free environment at a temperature of 400 to 800 °C [37]. Phenolic monomers, tar, and gaseous products are formed when lignin is dissolved in pyrolysis [37]; [38]. Product composition differs with parameters like reactor type, temperature, heating rate, and residence time [39]; [37]. High-rate pyrolysis achieves maximum liquid bio-oil yield with high phenolic contents through high heating rates and brief reaction times [38]; [39]. Still, a drawback of this process is the intricate product distribution and polymeric tar generation [39]; [39]. Additionally, pyrolysis bio-oil is not usually directly usable as fuel; thus, it needs subsequent processes like hydrogenation and purification [40]; [39]. In addition, cross-linked polymers produced from side reactions during pyrolysis can decrease efficiency [37].

3.2 Partial Pyrolysis of Lignin

Partial pyrolysis of lignin takes place at comparatively low temperature zones, i.e., between 200 to 300 °C, and is not concerned with the total depolymerization of lignin to its basic phenylpropanoid units [34]; [41]. In this process, only the ether linkages present in the lignin structure are partially broken, while the carbon-carbon bonds are subjected to no breakage. Therefore, lignin remains a long-chain oligomer, which prevents its complete conversion into monomeric forms. At lower temperature limits, i.e., up to 250 °C, pyrolysis is a partial depolymerization process where lignin molecules are stable, resulting in breakage of only weaker bonds like the β-O-4 ether linkages [41]. Partial pyrolysis differs greatly from pyrolysis in that it is highly stable in maintaining the carbon structure of lignin within this temperature range [37]. Specifically, during the process, the lignin polymer's phenylpropanoid unit produces low-molecular-weight aromatic products, while other components mostly stay the same [37]; [42]. Oligomers with phenolic compounds and trace amounts of gas are the main products of partial pyrolysis. During pyrolysis, lignin is partially broken down by the breaking of ether bonds β-O-4, α-O-4, which releases lignin oligomers. In contrast, the lignin backbone's integrity is maintained for the synthesis of elongation products with phenolic structures due to the non-breakage of carbon-carbon C-C bonds [38]; [42].

Partial pyrolysis at the lower end of the temperature range successfully preserves the structural integrity of the lignin carbon matrix compared to more severe thermal treatments. This method, in turn, yields low-molecular-weight products rather than terminating in complete depolymerization of lignin, thus protecting the vital phenylpropanoid components. The resulting liquid products are typically described as low-emission phenolic materials and oligomers. In this regard, ether bond cleavage, especially of the β-O-4 type, plays a central role in the formation of oligomeric structures [38]. In contrast, the integrity of carbon-carbon bonds stabilizes the aromatic backbone, leading to lignin derivatives with structured chemical characteristics. Additionally, the low-temperature approach offers economic and environmental advantages by conserving energy and restricting the production of by-products. This low-temperature pyrolysis enables lignin to retain its carbon skeleton in relation to higher severity thermal treatment [37]. Within the temperature range produced via this technique, the yield of water-soluble product like phenolic and low molecular weight aromatic compounds is high, without breaking down the carbon skeleton critically [37]; [38].

Lignin oligomer extraction is also encouraged by combining the biochemical process with the catalytic boost during this pyrolysis procedure where the pyrolytic decomposition is managed [38]. Intramolecular forces including the van der Waal forces and the hydrogen bonding also keep the lignin construction units tightly held together at these temperatures. Figure 2 below illustrates the construction for the depolymerized lignin in part with the structural transformations during the procedure indicated.

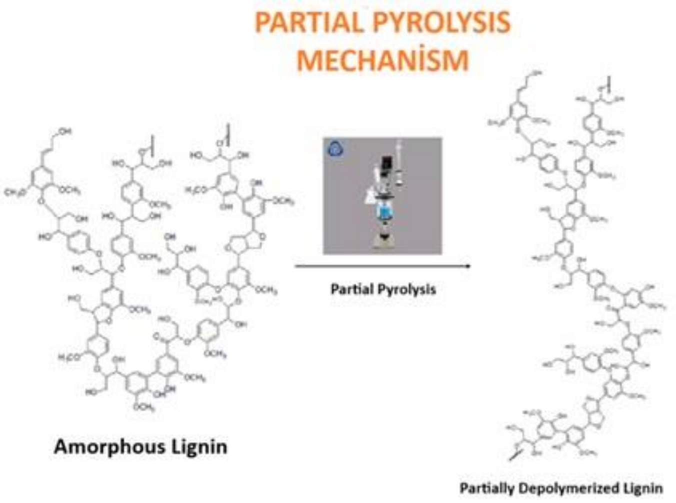

Figure 1. Formation of Partially Depolymerized Lignin

4. ADHESIVES

Wood adhesives were thoroughly discussed in the literature. The resin systems are typically categorized based on their chemical structure, and the most common class is formaldehyde F based systems. Adhesives in this class are urea UF, melamine MF, phenol PF and the MUF and PRF mixtures. But also highly reactive isocyanate-based adhesives are applied, e.g., polymeric diphenylmethane diisocyanate pMDI [43], emulsion polymer isocyanates EPI and polyurethane PUR systems [44]. Polyvinyl acetate PVAc and polyethylene-vinyl acetate EVA dispersions are common in the furniture and assembly industries. Biobased adhesives derived from tannin, protein or lignin are also significant options [45]. Natural polymers like tannin and lignin can be included in this category by reaction with formaldehyde [43].

Certain differences are made between structural binders e.g. for solid wood and wood composite board binders particleboard, MDF, OSB based on the application field. Categorize adhesives not only on the basis of the chemical basis but also on the basis of the chemical and mechanical response of the bond line into two main categories: in situ polymerized systems and pre-polymerized systems [44]. The first category includes rigid, highly cross-linked systems such as UF, MF, MUF, PF, PRF and pMDI.

The second group includes more flexible PVAc and PUR-based adhesives [44]. The differences between the two groups were stated to be very significant, especially for resisting moisture-induced stresses, which cause different types of failure .

4.1 Urea Formaldehyde Adhesives

Urea-formaldehyde UF resins are thermosetting adhesives that are extensively utilized in the production of wood-based composites and are known for their low price, rapid curing speed and superior bonding performance [46]; [47]. The resins form complex crosslinked three-dimensional networks via the condensation reaction of formaldehyde and urea to achieve superior bond strength [46]. Although UF adhesives are particularly favored in medium density fiberboard MDF, particleboard and plywood production; it is also of significant prominence in the construction and furniture industries because of the surface flatness benefit it offers [47]. Methylene bridges, which are the significant contributor to structure, are created through formaldehyde-urea reaction and enable quick curing even at low temperatures through acid catalysts [48]; [46]. The benefits of UF adhesives include low price, high bonding strength, and clear appearance after curing [49]. Because of these properties, UF adhesives are typically used in aesthetic surface applications, particularly in furniture production. Furthermore, the rapid curing rate speeds up the production processes and achieves economic efficiency through the saving of energy. But the most significant drawback of UF adhesives is that they are not resistant to water and moisture; therefore, they are not appropriate for outdoor use and their adhesive properties can weaken over time when exposed to humid environments. Formaldehyde, a hazardous and volatile chemical utilized in UF resins, is an important health and environmental hazard factor [50].

Specifically, long-term exposure has been linked to respiratory irritation, allergic reactions and possible carcinogenic effects. This prompted regulation of formaldehyde emissions with restrictions, and in this regard, studies on formaldehyde-free or low-emission adhesives as substitutes have intensified [51]; [50]. The emission of formaldehyde can be lowered by raising the urea/formaldehyde molar ratio, but at the expense of the bonding strength of the adhesive [52]. On the other hand, the emission of formaldehyde into the air can be avoided with the help of formaldehyde-absorbing additives. Low-emission UF adhesives have also been produced, and these adhesives have been designed to meet emission grades like E0 and E1. In addition, more robust and low-emission alternative resins, i.e., melamine formaldehyde MF and phenol formaldehyde PF adhesives, have also been created [53]. Although these alternatives are especially well-liked for exterior uses, bio-based adhesives i.e., soy-based adhesives also provide formaldehyde-free, eco-friendly alternatives [54]. The formaldehyde emissions can be lowered by raising the urea-formaldehyde U/F molar ratio but at the expense of the bond strength in urea-formaldehyde UF-bonded products [55]. The more realistic approach is to incorporate formaldehyde cleansers or absorbers e.g., tannins, lignosulfonates, or nano-fillers that bind chemically with free formaldehyde and lower its emission [56].

Meanwhile, new-generation low-emission UF resins are formulated to satisfy strict E0 and E1 levels of indoor air quality [57]. For outdoor or high-humidity applications, more durable resins like melamine-formaldehyde MF and phenol-formaldehyde PF show better moisture resistance at lower emissions [58]. Lastly, bio-based systems, specifically soy protein adhesives, have growing application as formaldehyde-free, environmentally friendly alternatives [59].

5. USE OF LIGNIN IN UF ADHESIVES

Lignin is a polyaromatic biomass by-product 50–75 million tons/year and the third most abundant natural polymer after cellulose and hemicellulose [56]; [60]. Due to the polyphenolic structure, it serves as an environmentally friendly alternative raw material for adhesives [60]. In resin uses, lignin has the potential to substitute urea or phenol partially or completely in thermosetting resins, thus saving petrochemical consumption of urea [56]. More significantly, lignin has many active hydroxyl and methoxyl groups which are capable of adsorbing free formaldehyde, thus acting as a formaldehyde emission reducer. For instance, adding modified lignin to UF resins has been demonstrated to greatly decrease formaldehyde emissions e.g., 74% decrease in a study and enhance indoor air quality [61]. Thus, lignin application is being explored with the objective of minimizing VOC and HCHO emissions from UF-bonded wood products. This strategy aids in attaining environmental objectives e.g., utilization of renewable waste materials and minimizes environmental and health problems [61]; [60].

Partial replacement of UF by lignin tends to decrease the quality of bonding and the strength of the panel onto which it is used, unless compensated for by formulation changes. Native lignin in UF resins reduces the bending strength MOR and internal bond IB values of particleboard and MDF [60]. For instance, bonded panels using neat lignin-based resins have had MOR values ranging from 8.1–11.0 N/mm² and IB values ranging from about 0.10–0.22 N/mm²; these are at the minimum necessary but lower than the case of UF being used individually reference MDF IB ~0.3–0.4 N/mm² [62]. Likewise, it has been noted that "panels manufactured with only UF resin have significantly better mechanical properties than panels bonded using lignin LS-UF adhesives". This is due to the lower cross-link density, as well as the larger molecular weight of lignin. Such is not the case, however, when additives or modification processes come into play. Inclusion of reactive crosslinkers or modification of lignin bonding mechanisms has a profound impact on performance. The addition of ~3% polymeric MDI pMDI to a 50:50 UF:Mg-lignosulfonate adhesive, for instance, increased IB by 366% from around 0.10 to 0.48 N/mm² and MOR by about 33% [60]. Additives UF + lignin + small pMDI can therefore restore and even surpass original strength. Moreover, nano-sized lignin additives can be utilized without strength loss. For instance, in a study, addition of 7% nanoparticles of dimethyl lignin to UF decreased HCHO but did not change/decrease the "physical and mechanical properties" of MDF [61]. In brief, the figures show that UF and pure lignin substitutions separately reduce MOR/IB but that such losses can be offset by the strategic application of partial substitutions like isocyanate additives or nano additives [60], [62].

Lignin addition effectively minimizes formaldehyde emission in UF panels [61]; [60]. In previous studies, the incorporation of lignin or lignosulfonate yielded far lower release than when UF was used by itself. For instance, MDF boards with 7% UF converter demethylated lignin nanoparticles released just 2.9 mg HCHO/100g formaldehyde. In boards made from UF resin alone in wood, there was a reduction by 74% from 11.2 mg/100g [61]. Wood panels with ammonium lignosulfonate and UF adhesive released low levels 0.7-1.0 mg/100g, as low as wood itself. That equates to a near-zero VOC [56]. To illustrate, adding Mg or Na lignosulphonate to UF was capable of decreasing formaldehyde by 91.1% and 56.9% respectively [60].

These values correspond to an E0 or super-E0 class emission ≤1.5 mg/100g, whereas the reference UF-bonded wood panel is an E1 ≤8 mg/100g Bekhta et al., 2021. These results confirm that lignin and lignin derivatives function as a scavenger of formaldehyde. The correct reactive position traps formaldehyde HCHO and minimizes the release. Quantitatively, the literature quotes 3-12 mg/100g for controls of lignin-modified UF wood panels [56]; [61]. This considerable reduction of formaldehyde release is also health- and environmentally friendly, rendering lignin-modified UF panels safe to be used indoors [56].

One of the most significant benefits of the application of modified lignin in UF resin is that the cost of raw materials is low. Lignin is an inexpensive and plentiful resource that comes from lignocellulosic material. It is used today as a fuel on a large scale and has been emphasized in the literature as being a "low-cost and widely used" additive [63]; [64]. However, the use of lignin often involves further processes demethylation, sulfonation, nanoparticle, pyrolysis, introducing some extra cost and process complexity in the production process [63]. In terms of processability, additives of lignin have great influence on resin properties. In a study, resins with sulfonated lignin content were found to be higher in pH, content of solids, viscosity and gel time but the formaldehyde release was reduced. Viscosity rose from 297.97 mPa-s to 4043.20 mPa-s and formaldehyde release reduced from 0.54% to 0.26% with an increase in the content of lignosulfonate from 0% to 2.5% [65]. This is because of crosslinking of lignin molecules in the adhesive and molecular weight increase. Consequently, lignin addition raises resin viscosity and strength and extends gel time and decreases formaldehyde release which can partly be compensated by pH adjustments. It has been found that lignosulfonate greatly improves the shelf life of UF resin. Lignosulfonate is said to enhance shelf life by enhancing electrostatic repulsion and lacks any chemical side reaction [64]. Chemical modification of lignin increases several parameters, namely resinoperability, which in turn affects its viscosity, pH value, and curing time and its long-term stability and formaldehyde emission. It is important to take into account viscosity-processing difficulties related thereto in using substantial levels of lignin. In short, empirical support exists for the inclusion of modified lignin in UF resins to provide numerous advantages in terms of environmental effect, human health, and sustainability. Additionally, urea-formaldehyde in conjunction with modified lignin can be used in a financially sound and aligned manner with the concept of resource conservation [63]; [64]. Free formaldehyde emission has greatly reduced and ensured functionality and wood-based mechanical properties. Medium Density Fiberboard MDF and plywood can be tailored and conform to EN E0/E1 requirements [61]; [64]. However, it should be considered that there will be increased viscosity due to the inclusion of lignin, longer curing time, and specific processing for lignin.

Research findings indicate that pre-pretreated lignin can be used as potential replacements for conventional urea-formaldehyde resin systems in wood-panel, medium-density fiberboard MDF, and domestic furniture applications [61]; [63]. This research falls into this category because they look towards replacements that are environment friendly and meet both ecologically and health-oriented requirements [61].

6. UF RESIN STUDIES WITH PARTIALLY PYROLYZED LIGNIN

Presence of phenolic hydroxyl OH functional groups in partially pyrolysed lignin is dominant in the alteration of urea-formaldehyde UF resin systems owing to participation of such functional groups in binding interactions with molecules of formaldehyde. Therefore, lignin is an active material that provides a urea-type interaction with molecules of formaldehyde. A comprehensive experimental investigation examined the effect of adding 4-8% partially pyrolysed ammonium lignosulphonate ALS to UF resin through differential scanning calorimetry DSC. Heat reaction was found to rise with an increase in lignin concentration at 8%. Further, the degree of polymerisation as well as crosslink density was found to rise with the addition of partially pyrolysed lignin. The reaction of polymerisation of formaldehyde with carbonyl or phenolic groups of the lignin structure leads to the decrease of free formaldehyde HCHO emissions [56] Therefore, the incorporation of partially pyrolysed lignin into the UF resin has the following effects:

i Partially pyrolysed lignin PPPL is a potential adsorbent for formaldehyde.
ii It assists in attaining greater network densities at low concentrations.
iii Without the presence of nitrifying ions, the accelerating effect of catalysis does not function. Beyond changing the curing kinetics, partially pyrolysed lignin addition to urea-formaldehyde adhesives brings an aromatic effect to the crosslinked structure [56].

The proportionate characteristics usually define the intensity of mechanical properties achieved from the application of UF resins with partially pyrolysed lignin. UF resin composite panels made fully from UF resin have mechanical properties that are marginally superior to those produced through the integration of UF resins and unmodified lignin. For example, it was reported in one study that the flexural strength MOR of a wood panel consisting of UF resin was about 4.88 MPa and its modulus of elasticity MOE was 573 MPa. When, however, a resin with 75% pyrolysis oil content 3:1 PyO:UF was used, the IB value reduced from 0.101 to 0.062, the MOR reduced to 2.30 MPa, and the MOE reduced to 189 MPa. The MOR of the board on 50% 1:1 based resin in the same study was around 0.64 MPa, a low value displaying a 67% decrease [66]. In other words, high lignin/oil content reduces the internal bond strength and reduces the internal bond homogeneity of the panel. Conversely, another study demonstrated that experiments with partially pyrolyzed lignosulfonate-based resins showed that the panels made with resins possessing 20% lignin achieved superior mechanical properties: MOR 15 MPa, MOE 3320 MPa, IB 0.48. These values show the partially pyrolyzed lignin meets the mechanical test standards. In conclusion, the inclusion of pyrolyzed lignin usually decreases mechanical properties for high addition ratios, etc., and although partially pyrolyzed lignin, given the appropriate addition ratios can provide enough strength. The numerical examples we have demonstrated state the following: UF without additives: MOR 4.9 MPa, IB 0.10; High-content pyrolyzed lignin 3:1, MOR 2.3 MPa, IB 0.062 approximate 50% reduction [66]; Partially pyrolyzed lignosulfonate 20%, MOR 15 MPa, IB ~0.48 [62]. The data demonstrate that their very well may be losses or improvements in sheet strength depending on lignin addition dosage.

Our research; looked in depth into the chemical, mechanical, and environmental performance of new-generation adhesives, which included UF resin systems that have been modified using partially pyrolyzed lignin. The literature shows that the partial pyrolysis of lignin at mild temperatures 150–300 °C produces low molecular weight, reactive lignin forms that still have phenolic structures. These lignin forms provide chemical bonding contributions and environmental benefits to UF resin systems. In the reaction of partially pyrolyzed lignin with formaldehyde, the aromatic –OH groups in the lignin structure can undergo condensation reactions with formaldehyde to capture free formaldehyde. The numerical data set forth in the studies demonstrate that, in panels, formaldehyde emissions can be reduced to 1.4–2.0 mg/100 g by including 4–8% partially pyrolyzed lignin, which corresponds to the E0 class for formaldehyde emissions. The reduction of formaldehyde is imperative because high indoor concentrations of formaldehyde and related emissions can reduce indoor air quality by making occupants ill.

In the case of mechanical performance, the study noted that for UF resins with the addition of partially pyrolyzed lignin, did not negatively influence mechanical properties IB, MOR, and MOE even when low amounts were added and may influence these properties in some situations positively. However, increasing the amount of lignin can lead to effects like increased viscosity of the resin, an increase in the cure time, and a reduction in wood panel strength. Therefore, a compromise is necessary between the performance and the lignin addition rate. Partially pyrolyzed lignin-based modification importantly has sustainability implications. Lignin is a waste product from the paper and bioethanol industries used as a low-value fuel; but it is possible to reprocess lignin through partial pyrolysis and make high-value-added products from it. The addition of partially pyrolyzed lignin may help reduce fossil raw material in resin systems, will lower the carbon footprint, and supports a renewable resource production model.

That said, the research in this area is somewhat underwhelming. There have been, especially, very few studies on the use of partially pyrolyzed lignin form in UF resins. There are many reasons for this including the heterogeneous nature of partially pyrolyzed products, the diverse sources of lignin, the inability to standardize pyrolysis conditions, and limited understanding of interaction mechanisms with resins which certainly justify further experimental and theoretical investigations in this area. In summary, partially pyrolyzed lignin has great potential to not only decrease formaldehyde emissions, but also to improve properties and enhance the percentage of bio-based content in UF resins. Yet, these systems will only continue to become viable alternatives if we develop additional literature data, scalable process development, and methods of testing and standardization. Furthermore, UF resin systems modified with partially pyrolyzed lignin represent a novel transition that could lead to an eco-friendly transition in the wood-based materials industry and help develop sustainable adhesive technology.

7. CONCLUSION

The current research examined comprehensively the chemical, mechanical, and environmental characteristics of novel adhesives from UF resin systems modified with partially pyrolyzed lignin. The literature data indicate that the low-temperature pyrolysis 150–300 °C of lignin yields low molecular weight, reactive derivatives of phenolic structure that are preserved. Chemical bonding contributions and environmental benefits are both present in lignin derivatives in UF resin systems.

Under the pyrolysis of lignin in the presence of formaldehyde, the aromatic –OH groups in the lignin molecule become condensed with formaldehyde, encapsulating free formaldehyde. According to the quantitative data presented in the investigations, formaldehyde emissions from the panels can be reduced to 1.4–2.0 mg/100 g with the addition of 4–8% partially pyrolyzed lignin, comparable to the E0 class. This is particularly beneficial for improving indoor air quality and reducing harmful emissions that are not good for the health of human beings. In mechanical performance, the addition of partially pyrolyzed lignin was found not to have a negative effect on the mechanical properties of UF resins such as internal bond strength IB, bending strength MOR, and elastic modulus MOE, even with low levels of additions, and in some cases may even increase these values. But to raise the addition rate can have the following implications such as increased viscosity of the resin, increased curing time, and reduced strength in the wood panel. Again, here there must be a trade-off between lignin addition rate and performance. Modification of the partially pyrolyzed lignin is also important on the sustainability side. Lignin is a byproduct in paper and bioethanol production and is used as a low-value fuel but can be recycled through partial pyrolysis and converted to high-value-added products. Through the addition of partially pyrolyzed lignin, use of fossil raw materials in resin systems is reduced, the carbon footprint is reduced, and a model for production based on renewable resources is promoted. But few studies exist in this field of literature. Particularly, few studies are available on the utilization of partially pyrolyzed lignin in UF resins. Structural heterogeneity in partially pyrolyzed products, diversity of lignin sources, lack of standardization in pyrolysis conditions, and incompleteness of knowledge in understanding interaction mechanisms with resins render experimental and theoretical studies in this field imperative.

In conclusion, partially pyrolyzed lignin has great potential to reduce the emission of formaldehyde, increase mechanical properties, and increase the bio-based content ratio in UF resins. For these systems to become a commercially acceptable alternative, additional literature data, scale-up process development, and standardization studies are needed. In this sense, UF resin systems formulated using partially pyrolyzed lignin seem to be a new area of research that can spark an environmental revolution in the wood-based materials industry and contribute towards the development of environmental friendly adhesive technology.

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Authors' Contributions

No	Full Name	ORCID ID	Author’s Contribution
1	Hande Yılmaz	0009-0004-9577-323X	1,2,3,4
2	Doğu Ramazanoğlu	0000-0002-6356-5792	1,2,4
3	Halil İbrahim Uğraş	0000-0002-1633-967X	2,3,4
4	Ayhan Tozluoğlu	0000-0002-1828-9450	1,3,4
1-Structural Revision, 2-Literature Inclusion, 3-Methodology 4-Guidance for Future Studies			

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