

Article

Urea–Formaldehyde Synthesis with Kraft Lignin Nanoparticles and Effects on Formaldehyde Emissions

Thamirys Andrade Lopes ¹, Angélica de Cássia Oliveira Carneiro ¹, Thaynara Silva Vieira ¹, Gustavo Henrique Denzin Tonoli ², Iara Fontes Demuner ^{1,*} , Marcela Ribeiro Coura ¹, Verônica Oliveira de Paula Barbosa ¹ , Ana Márcia Macedo Ladeira Carvalho ¹ , Letícia Costa Peres ¹ , Tafla Veloso de Oliveira ³ , Fernando Mata ^{3,4} , Joana Santos ³  and Meirielly Jesus ^{3,*} 

¹ Department of Forestry Engineering, Federal University of Vicosa, Av. Peter Henry Rolfs, Vicosa 36570-900, Brazil; thamiryscpo@gmail.com (T.A.L.); cassiacarneiro@ufv.br (A.d.C.O.C.); ana.marcia@ufv.br (A.M.M.L.C.)

² Department of Forest Sciences, Federal University of Lavras, Trevo Rotatório Professor Edmir Sá Santos, Lavras 37203-202, Brazil

³ CISAS—Center for Research and Development in Agrifood Systems and Sustainability, Instituto Politécnico de Viana do Castelo, 4900-367 Viana do Castelo, Portugal

⁴ Estação Zootécnica Nacional, Instituto Nacional de Investigação Agrária e Veterinária (INIAV), 2784-424 Vale de Santarém, Portugal

* Correspondence: iara.demuner@ufv.br (I.F.D.); meiriellyjesus@ipvc.pt (M.J.)

Abstract

To seek improvements in the quality and sustainability of adhesives for bonding wood, the objective of this work was to use pine and eucalyptus kraft lignin nanoparticles in the synthesis of urea–formaldehyde adhesives. The rheological properties, shear strength of glued joints and formaldehyde emission reduction were evaluated. The lignin nanoparticles were produced from pine and eucalyptus kraft lignins, and two milling conditions were evaluated, in which the material was passed through the mill 5 and 10 times. The adhesives were synthesized by adding 0, 2, 4, 6, 8 and 10% lignin nanoparticles, replacing the total mass of urea. The treatments with better performances in relation to the control were submitted to Fourier transform infrared (FTIR) analysis and quantification of formaldehyde emissions. The lignin nanoparticle addition promoted an increase in the viscosity and pH of the adhesives. The treatments with 4% eucalyptus lignin nanoparticles (E5P 4%) and 6% pine lignin nanoparticles (P5P 6%) increased the shear strength by 10% and 13% in the dry and wet conditions, respectively. At higher replacement levels, there was a decrease in resistance. An increase in the intensity of the peaks at 1240 cm^{−1} (C–N stretching and angular deformation of NH₂) and 1031 cm^{−1} (stretching of C–N bonds or N–C–N of methylene-ether bonds) was observed. The formaldehyde emissions of selected adhesives were reduced by up to 75%. The E5P 4% and P5P 6% adhesives had the best performances. Lignin nanoparticles in the urea–formaldehyde adhesive promoted improvements in the quality of the adhesive and replacement of urea by a renewable resource. Another highlight is the sustainability, due to the reduction in formaldehyde emissions obtained in adhesives produced with lignin nanoparticles.



Academic Editor: Fábio Fernandes

Received: 20 May 2026

Revised: 3 July 2026

Accepted: 5 July 2026

Published: 9 July 2026

Copyright: © 2026 by the authors.

Licensee MDPI, Basel, Switzerland.

This article is an open access article distributed under the terms and conditions of the [Creative Commons Attribution \(CC BY\)](https://creativecommons.org/licenses/by/4.0/) license.

Keywords: sustainability; adhesive synthesis; formaldehyde emission; kraft lignin nanoparticles

1. Introduction

The performance of adhesive bonds between wood components depends on the physicochemical characteristics of both the adhesive and the wood, as well as the processing

parameters employed during bonding, the geometry and size of the bonded elements, and the service conditions to which the glued wood is exposed [1,2]. Consequently, the stability and useful life of bonded wood products are strongly influenced by the strength, stiffness, and durability of the adhesive joints [3].

Urea–formaldehyde adhesives are widely used in the manufacture of plywood, particleboard, medium-density fiberboard, and other wood-based panels because of their low cost, fast curing, and good bonding performance under dry conditions [4,5]. However, compared with phenol–formaldehyde adhesives, they exhibit lower moisture resistance and release higher levels of formaldehyde.

Reducing formaldehyde emissions by lowering the formaldehyde-to-urea molar ratio may adversely affect the curing behavior, crosslink density, and bonding strength [6,7]. Likewise, incorporating additional components into the resin formulation may reduce formaldehyde emissions but can increase viscosity and impair resin penetration into the wood substrate. Therefore, urea-based adhesives require modifications to reduce formaldehyde emissions without compromising their mechanical bonding performance.

To improve the quality and performance of these adhesives, alternative renewable materials have been evaluated. Kraft lignin is a major by-product of the kraft pulp production process and is currently used primarily as a fuel for energy generation. However, within the biorefinery concept, its application in high-value products has attracted attention as a strategy to enhance the sustainability of kraft mills through more efficient utilization of lignin [8–12].

Kraft lignin is a biodegradable, environmentally friendly, renewable, and sustainable material, which encourages its use in polymers [13]. Moreover, it contains aromatic structures, phenolic and aliphatic hydroxyl groups, and other reactive sites that may interact with formaldehyde or participate in the crosslinked network of urea-based adhesives [14]. The chemical structure and reactivity of lignin vary depending on the feedstock. Softwood lignin (e.g., pine) is composed mainly of guaiacyl (G) units, whereas hardwood lignin (e.g., eucalyptus) contains both guaiacyl (G) and syringyl (S) units [15]. These structural differences may influence the degree of condensation, hydroxyl availability, steric accessibility, and reactivity toward formaldehyde during adhesive synthesis.

Another factor affecting lignin reactivity in adhesive synthesis is particle size. Kraft lignin typically has particle sizes ranging from 10 to 100 μm , which may hinder its dispersion in polymer matrices [16,17] and reduces its reactivity toward formaldehyde [14,18–20]. Therefore, strategies to increase its reactivity have been investigated, including the production of lignin nanoparticles.

Lignin nanoparticles can be produced by a mechanical process using kraft lignin [21–23]. Compared with chemical methods, physical–mechanical approaches are generally less expensive, require no strict temperature control, and do not generate toxic or hazardous substances [24].

The use of lignin at the nanoscale increases its specific surface area, facilitating dispersion and incorporation into polymer matrices. In addition, lignin nanoparticles retain a variety of reactive functional groups, such as thiols, alcohols and aliphatic and phenolic hydroxyl groups, broadening their potential applications [25].

The incorporation of lignin nanoparticles into adhesives has the potential to improve the physical, mechanical, and thermal properties of biocomposites [23]. Most studies have focused on adding lignin nanoparticles to commercial phenol–formaldehyde adhesives or partially replacing phenol–formaldehyde during their synthesis. Chen et al. [26] reported improvements in adhesive performance, particularly in mechanical strength.

However, studies on the synthesis and modification of urea–formaldehyde adhesives using lignin nanoparticles remain limited, and the influence of lignin nanoparticles on the

adhesive structure, curing behavior, and bonding performance is not yet fully understood. Furthermore, the effects of the lignin nanoparticle source (pine or eucalyptus) and incorporation level on the synthesis, properties, and performance of urea–formaldehyde adhesives have not been systematically investigated.

The incorporation of lignin nanoparticles is expected to improve the performance of urea–formaldehyde adhesives, particularly by enhancing moisture resistance and reducing formaldehyde emissions. Their hydrophobic character and high specific surface area may promote interactions with urea–formaldehyde molecules, reducing the availability of free hydroxymethylol groups and limiting the hydrolytic degradation of aminomethylene linkages after curing [27]. In addition, lignin nanoparticles may act as formaldehyde scavengers by reacting with residual free formaldehyde before or during curing.

The reduction in formaldehyde emissions is of great importance for the wood adhesive and panel industries because formaldehyde is classified as a Group 1 carcinogen by the International Agency of Research on Cancer (IARC) [28] and is known to cause irritation of the eyes, skin, and respiratory tract [29–31]. Therefore, the objective of this study was to evaluate the use of kraft lignin nanoparticles from pine and eucalyptus in the synthesis of urea–formaldehyde adhesives for wood bonding, aiming to improve the quality and sustainability of the adhesive and wood panel sector.

2. Materials and Methods

Powdered kraft lignins from *Eucalyptus* sp. and *Pinus* sp., obtained from industrial sources and isolated by acid precipitation, were used in this study. After isolation, the lignins were conditioned in a climate-controlled room at 23 ± 1 °C and $50 \pm 2\%$ relative humidity for 48 h before milling. The eucalyptus kraft lignin exhibited a moisture content of 9.0%, an ash content of 0.32%, a sulfur content of 1.97%, and a pH of 2.1. In contrast, the pine kraft lignin exhibited a moisture content of 9.3%, an ash content of 2.37%, a sulfur content of 3.50%, and a pH of 3.2.

2.1. Lignin Nanoparticle Production

Kraft lignin nanoparticles from *Eucalyptus* sp. and *Pinus* sp. were produced by a mechanical process using a Super Masscolloider Masuko Sangyo mill (MKCA6-2, Msuko Sangyo Co., Kawaguchi, Japan) operating at 1500 rpm. The specific energy consumption of 1.0×10^4 kWh/ton was obtained directly from the equipment. The oven-dry lignin content was determined before suspension preparation, and lignin suspensions were prepared in distilled water at a consistency of 5% (*w/v*), without pH adjustment and without the addition of any dispersant, surfactant, or stabilizer. The distance between the grinding disks was set to 100 µm, as described by Wang et al. [32].

For each lignin type, two processing levels corresponding to the number of passes through the mill (5 and 10 passes) were evaluated. The milling process was performed in duplicate. The lignin nanoparticle preparation times were 45 min and 90 min for 5 and 10 passes, respectively.

After milling, the suspensions were centrifuged at 4000 rpm for 60 min at 10 °C to remove excess water. The supernatant was discarded, and the recovered lignin nanoparticle precipitate was transferred to trays and rapidly frozen at -80 °C for 24 h in an ultra-low-temperature freezer. The frozen samples were subsequently freeze-dried for 48 h using an Enterprise I freeze dryer under a vacuum of 160 µHg with the condenser maintained at -45 °C to remove residual water. After freeze-drying, the samples were stored in sealed plastic containers to prevent moisture absorption prior to further analyses.

2.2. Lignin Nanoparticle and Kraft Lignin Characterization

The morphologies of kraft lignins and lignin nanoparticles were obtained from scanning electron microscopy (SEM) and transmission electron microscopy (TEM), respectively. For SEM analysis, lignin samples were dispersed in deionized water at a concentration of 0.1 mg mL^{-1} and sonicated for 10 min. A drop of lignin suspension was deposited onto an aluminum stub and dried at room temperature. The samples were sputter-coated with an approximately 10 nm thick gold layer using a Quorum Q150RS sputter coater under an argon atmosphere at a pressure of 0.5 bar. SEM observations were performed using a JEOL JSM-IT700HR microscope (JEOL Ltd., Tokyo, Japan) at a working distance of approximately 10 mm and magnifications ranging from $5000\times$ to $50,000\times$. At least five representative images were collected from different sample regions, and micrographs exhibiting the predominant particle morphology and free of preparation artifacts were selected for analysis, following the procedure described by Mishra and Ekielski [33].

For TEM analysis, a drop of the lignin nanoparticle suspension was deposited onto carbon-coated copper grids and allowed to dry at room temperature for 24 h prior to analysis. TEM images were acquired using a Zeiss EM 109 transmission electron microscope (Carl Zeiss AG, Baden-Württemberg, Germany). operated at an accelerating voltage of 120 kV. At least five representative micrographs were recorded from different regions of the grid for morphological evaluation. The average nanoparticle diameter was determined by digital image analysis using ImageJ software (version 1.54, National Institutes of Health, Bethesda, MD, USA) based on measurements of 100 randomly selected particles from the representative TEM micrographs. Before analysis, the images were calibrated using the microscope scale bar, and the diameter of each particle was measured along its longest axis. The resulting measurements were used to calculate the average particle size and particle size distribution.

The chemical functional groups present in each type of lignin and the lignin nanoparticles were characterized by Fourier transform infrared (FTIR) spectroscopy using a Varian 660-IR spectrometer equipped with an attenuated total reflectance (ATR) accessory (GladiATR, PIKE Technologies, Fitchburg, WI, USA) fitted with a diamond crystal. Spectra were recorded in the range of $400\text{--}4000 \text{ cm}^{-1}$ at a spectral resolution of 4 cm^{-1} , using 32 scans per spectrum to improve the signal-to-noise ratio. Before each measurement, a background spectrum was collected under identical conditions and automatically subtracted from the sample spectra. All spectra were processed using Origin software (OriginPro 2023b version, OriginLab Corporation, Northampton, MA, USA). For each sample, measurements were performed in triplicate to ensure reproducibility, and the average spectrum was used for analysis.

The kraft lignins and lignin nanoparticles used in this study were obtained from the same production batches previously characterized by Lopes et al. (2020) [34]. Therefore, the compositional data reported in that study are representative of the materials evaluated here. For completeness, the main compositional results are summarized as follows: the total lignin contents were 95.98% for pine kraft lignin and 93.81% and 92.70% for the corresponding lignin nanoparticles produced after 5 and 10 milling passes, respectively. For eucalyptus kraft lignin, the total lignin content was 96.89%, while the lignin nanoparticles produced after 5 and 10 milling passes exhibited total lignin contents of 95.53% and 95.64%, respectively. No additional compositional analyses were carried out in the present study because the same material batches were used throughout all experiments.

2.3. Adhesive Synthesis

The synthesis of the urea–formaldehyde control adhesive was based on the methodology by Ferreira (2017) [1]. Formaldehyde (molecular weight: 30.03 g/mol) diluted to 37%,

powdered urea (molecular weight: 60.06 g/mol), sodium hydroxide (NaOH) diluted to 20% and formic acid diluted to 30% were used.

An initial formaldehyde:urea (F:U) molar ratio of 2.09:1 was considered to obtain a final molar ratio of 1.26:1. Amounts of 170 g of formaldehyde and 60 g of urea were added to a 500 mL flat-bottomed tritube flask. This mixture was taken to a heating plate with constant mechanical agitation, in a laminar flow hood, equipped with a condenser and thermometer. The solution was heated to 90 °C. The pH of the solution was adjusted to 9.0 using NaOH, maintaining heating at a temperature of 90 °C for 45 min.

After this time, the pH of the solution was adjusted to 5.0 with formic acid. The reaction continued at 90 °C for 15 min. Then, 30 g of urea was added to the solution, and the temperature was maintained at 90 °C for another 20 min. The pH of the mixture was adjusted to approximately 8.0, with the addition of NaOH, and another 10 g of urea was added to the solution. In this synthesis step, the temperature was reduced to 70 °C and the reaction maintained for 45 min. After this period, the synthesis was completed, and the adhesive was cooled to room temperature.

For the lignin nanoparticle–urea–formaldehyde adhesives, the lyophilized lignin nanoparticles were added in percentages of 2, 4, 6, 8 and 10%, replacing the total mass of urea to be used in the synthesis, added at the beginning of the synthesis. Table 1 presents the exact masses of the nanoparticles used in the adhesive formulations. It should be noted that sodium hydroxide (NaOH) and formic acid were used only for pH adjustment.

Table 1. Exact masses of urea, formaldehyde solution, and lignin nanoparticles used in adhesive formulations.

	0%	2%	4%	6%	8%	10%
Urea (g)	60 + 30 + 10	58.8 + 30 + 10	57.6 + 30 + 10	56.4 + 30 + 10	55.2 + 30 + 10	54 + 30 + 10
Formaldehyde (g)	170	170	170	170	170	170
Lignin nanoparticles (g)	-	1.2	2.4	3.6	4.8	6.0

2.4. Adhesive Characterization

The adhesive formulations were characterized according to their solid content [35], pH [36], and viscosity [37].

The shear strength of the glue line under dry and wet conditions was determined according to ASTM D2339-98 (1998b) and ABNT NBR ISO 12466-1 (2006) standards, respectively [38,39]. For these tests, two-plywood was prepared from *Pinus* sp. veneers with a basic density of 0.490 g cm⁻³ and a shear strength of 6.91 MPa at 12% moisture content. The veneers measured 0.65 × 9.0 × 40.0 cm (thickness × width × length). A single glue line was produced using a double-sided adhesive application at a spread rate of 250 g m⁻². The veneers were assembled immediately after adhesive application (open assembly time ≈ 0 min), followed by cold pre-pressing for 4 min (closed assembly time). Subsequently, the panels were hot-pressed in a hydraulic press at 180 °C under a pressure of 12 kgf cm⁻² for 8 min. After pressing, the two-plywood was conditioned in a climate chamber at 20 ± 2 °C and 65 ± 3% relative humidity until constant mass was reached. Four laminated wood panels were produced for each treatment, and the shear test specimens were subsequently prepared in accordance with the requirements of the respective ASTM and ABNT standards. The wet shear strength was determined according to the procedures specified in ABNT NBR ISO 12466-1 [39], after immersing the specimens in water at 20 ± 3 °C for 24 h. The specimens were tested immediately after removal from the water, in accordance with the standard. Shear strength tests were performed using a universal testing machine (Model I4230, Contenco Indústria e Comércio Ltda., Minas Gerais, Brazil).

equipped with a 15 kN load cell. The tests were conducted at a crosshead speed of 6 mm min⁻¹. The percentage of wood failure was not evaluated in this study.

For adhesive samples, FTIR-ATR analyses were carried out after curing under the conditions described in Section 2.2.

The formaldehyde emission test was performed following the procedures described in the ASTM (2014) standard, which uses the desiccator method [40]. Quantification was obtained by preparing solutions containing chromotropic acid, in a strongly acidic medium, according to ASTM (2014) [40].

2.5. Experimental Design and Statistical Analyses

The experiment was conducted using a completely randomized design in a factorial arrangement consisting of two lignin nanoparticle types (Pinus and Eucalyptus), two lignin nanoparticle production levels (5 and 10 milling passes), and five freeze-dried lignin nanoparticle concentrations (2, 4, 6, 8, and 10%), plus a control treatment (0%), resulting in 21 treatments with three replicates each, for a total of 63 experimental units.

The data were subjected to analysis of variance (ANOVA), and when significant effects were detected, treatment means were compared using Tukey's test at the 5% significance level. For quantitative variables, regression models were fitted whenever appropriate, based on the significance of the regression coefficients, coefficients of determination (R^2), standard error of the estimate (SE), and residual analyses. In addition, the treatments were compared with the control using Dunnett's test ($p < 0.05$).

To select the adhesive formulations for Fourier transform infrared spectroscopy (FTIR) and formaldehyde emission analyses, hierarchical cluster analysis based on Euclidean distance was performed using the viscosity, dry shear strength, and wet shear strength as the clustering variables.

For formaldehyde emissions, the data were analyzed by ANOVA, and treatment means were compared using Tukey's test ($p < 0.05$) whenever significant differences were observed.

3. Results and Discussion

3.1. Characterization of Kraft Lignins and Lignin Nanoparticles

The FTIR spectra of lignins and lignin nanoparticles from pine (A) and eucalyptus (B) are shown in Figure 1. For each wood species, the spectra are highly similar, indicating that the mechanical milling process did not induce major chemical changes detectable by FTIR under the conditions evaluated. Since the spectra were not normalized, the discussion is based on the presence and assignment of the absorption bands rather than on comparisons of their relative intensities.

The broad absorption band between 3500 and 3100 cm⁻¹ is attributed to the intramolecular stretching vibration of aromatic and aliphatic hydroxyls (-OH) groups [41]. The bands between 2900 and 2800 cm⁻¹, together with the band at 1455 cm⁻¹, are assigned to the C-H stretching and deformation vibrations of CH₂, CH₃, and OCH₃ groups [42,43].

The peak at 2350 cm⁻¹ is related to the stretching of the carboxyl groups (C=O), corresponding to the ketone group [39], while the absorption near 2150 cm⁻¹ has been attributed to C≡C stretching vibrations of acetylene compounds [44]. The absorption bands between 1700 and 1740 cm⁻¹ correspond to the stretching vibrations of carbonyl (C=O) groups associated with non-conjugated ketones and carboxylic acids [41,42]. The region between 1600 and 1400 cm⁻¹ is characteristic of aromatic skeletal vibrations resulting from C=C stretching in the lignin aromatic rings [45–47], whereas the band at 1370 cm⁻¹ is assigned to the in-plane deformation of phenolic hydroxyl groups [42].

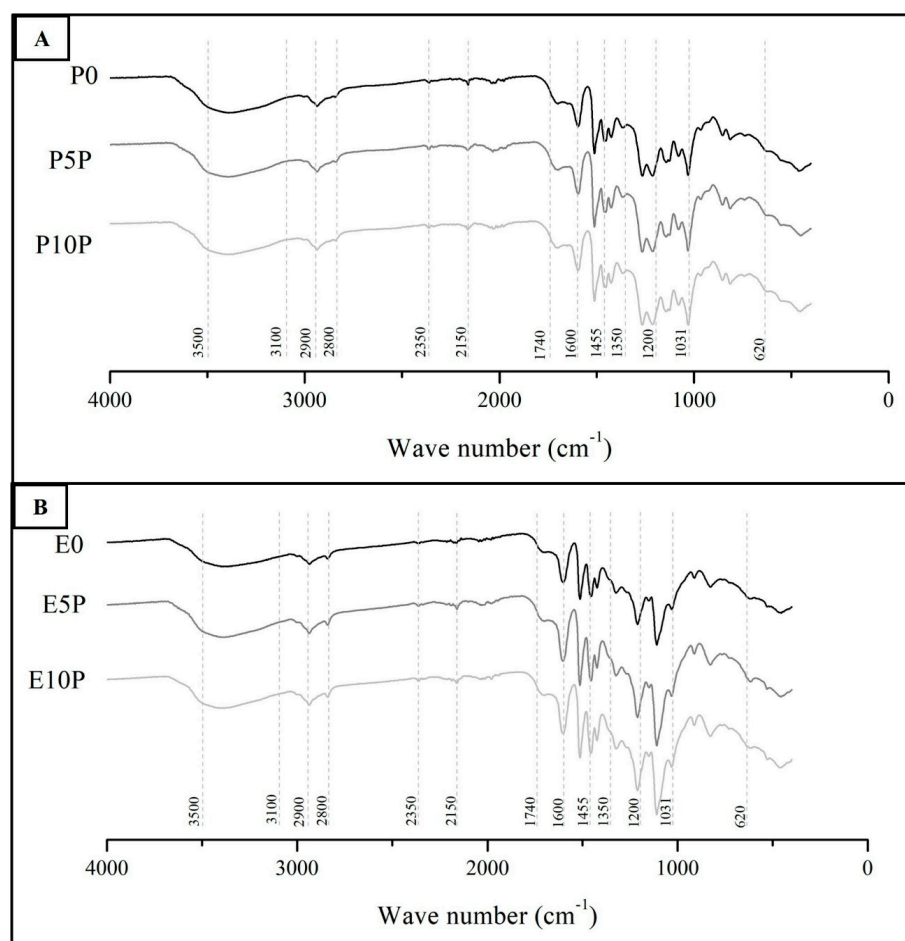


Figure 1. FTIR of lignins and lignin nanoparticles from pine (A) and eucalyptus (B). P0 (pine lignin); P5P (pine lignin nanoparticles: 5 passes); P10P (pine lignin nanoparticles: 10 passes); E0 (eucalyptus lignin); E5P (eucalyptus lignin nanoparticles: 5 passes); E10P (eucalyptus lignin nanoparticles: 10 passes).

Absorption bands were observed in the range of 1300 cm^{-1} and 1200 cm^{-1} , which are assigned to guaiacyl and syringyl units, respectively. These bands have also been reported by Lopes (2018) and Watkins et al. (2015) [34,48]. As expected from their botanical origin, eucalyptus lignin exhibited bands associated with both guaiacyl and syringyl units, whereas pine lignin predominantly exhibited bands characteristic of guaiacyl structures. The absorption observed near 1200 cm^{-1} in pine lignin may also be related to O–H bending and aromatic C–O stretching vibrations [46,48].

The band at approximately 1050 cm^{-1} is attributed to alkyl ether ($\beta\text{--O--4}$) linkages [43], while the band at 1031 cm^{-1} is associated with in-plane aromatic C–H deformation and C–O stretching vibrations of primary alcohols [48]. The absorption band at approximately 620 cm^{-1} has been attributed to sulfur-containing groups originating from the kraft pulping process [49].

The presence of phenolic hydroxyl groups and aromatic structures may enhance the interactions between lignin/lignin nanoparticles and the urea–formaldehyde matrix, strengthening intermolecular bonding and potentially improving mechanical properties while reducing formaldehyde emissions. In addition, the higher content of carboxylic acids and phenolic groups may increase lignin reactivity, promoting a greater extent of interactions and possible crosslinking with the urea–formaldehyde matrix [50], particularly through hydrogen bonding and condensation reactions involving available functional groups. Together, these functional groups suggest that lignin/lignin nanoparticles may act

not only as a passive filler but also as reactive components capable of participating in the adhesive network, thereby influencing the adhesive performance.

SEM and TEM micrographs (Figure 2) show the reduction in the particle sizes of pine and eucalyptus lignins from the microscale to the nanoscale.

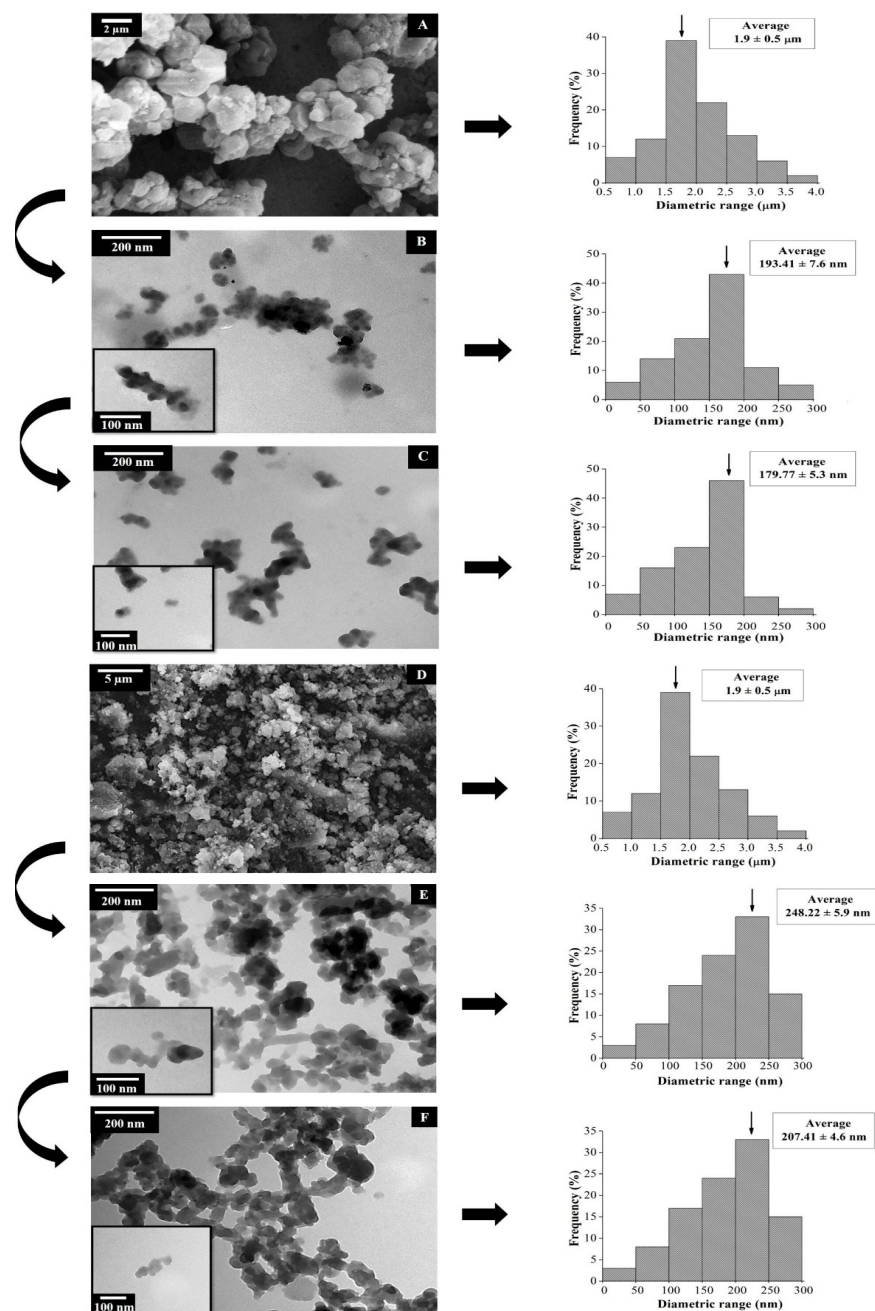


Figure 2. SEM and TEM micrographs of eucalyptus and pine lignins and lignin nanoparticles and their respective diameter frequencies. SEM of eucalyptus (A) and pine (D) lignin particles; TEM of eucalyptus (B) and pine (E) lignin nanoparticles; TEM of eucalyptus (C) and pine (F) lignin nanoparticles after 10 passes in the mill.

According to the SEM images, the eucalyptus (A) and pine (E) kraft lignins consisted of irregular shaped particles with a broad size distribution in the micrometer range. Analyzing the TEM micrographs revealed the presence of isolated nanoparticles with diameters below 100 nm and a predominantly spherical morphology for both lignin nanoparticle types. These nanoparticles tend to form agglomerates of different shapes, with average

sizes around 200 nm. Similarly, Nair et al. (2014) and Lopes et al. (2018), using mechanical methods to obtain lignin nanoparticles (mechanical homogenization and SuperMassColloder mill, respectively), reported particles in the nanometer range with a pronounced tendency to agglomerate [34,51].

The mean particle sizes of eucalyptus lignin after 5 and 10 passes through the grinder were 193.41 nm and 179.77 nm, respectively, corresponding to a reduction of 13.64 nm. For pine lignin, the mean particle sizes after 5 and 10 passes were 248.22 nm and 207.41 nm, respectively, representing a reduction of 40.81 nm. Tukey's test at the 5% significance level revealed statistically significant differences between the particle sizes obtained after 5 and 10 passes for both lignin types, confirming that increasing the number of milling passes promoted additional particle size reduction. However, the magnitude of the reduction differed between the lignins, being substantially greater for pine lignin than for eucalyptus lignin.

TEM observations indicated that both milling conditions produced nanoparticles with a tendency to agglomerate. Nevertheless, no clear increase in the degree of agglomeration was observed when the number of milling passes increased from 5 to 10. Therefore, under the processing conditions evaluated in this study, 10 milling passes significantly reduced the particle size without causing noticeable changes in the particle morphology or agglomeration.

Eucalyptus lignin nanoparticles exhibited smaller and more homogeneous particles, whereas pine lignin nanoparticles formed larger and more irregular agglomerates. These differences may be related to the intrinsic structural characteristics of the parent lignins. Hardwood lignins, such as eucalyptus, contain both syringyl (S) and guaiacyl (G) units and generally exhibit a lower degree of condensation than softwood lignins, which are predominantly composed of guaiacyl units [15].

Thus, the higher condensation degree typically observed for pine kraft lignin increases its mechanical resistance, favoring the formation of larger particles and aggregates after milling. In addition, the higher ash content of pine kraft lignin may also contribute to this behavior. Consequently, the smaller particle size, more homogeneous morphology, and lower ash content of eucalyptus lignin nanoparticles may facilitate their dispersion within the urea–formaldehyde resin matrix, thereby increasing the available surface area for interactions with the polymer network.

3.2. Characterization of Adhesives Synthesized with Pine and Eucalyptus Lignin Nanoparticles

The pH values of the adhesives as a function of the addition of lignin nanoparticles (2, 4, 6, 8 and 10%) are shown in Figure 3.

The pH of the adhesives increased with the increasing lignin nanoparticle content. During adhesive synthesis, the same formaldehyde-to-urea molar ratio used for the control formulation was maintained, while lignin nanoparticles were incorporated by partially replacing the urea mass. This approach was adopted because the molecular weight and polydispersity of the lignin nanoparticles were not sufficiently defined to allow for stoichiometry adjustment.

In the final stage of the synthesis, an additional amount of urea was added to react with the remaining free formaldehyde, forming methylolurea species [51]. As the lignin nanoparticle content increased, a greater proportion of the free formaldehyde may have reacted with the reactive sites of lignin, reducing the amount of formaldehyde available to react with the final urea addition. Consequently, part of the added urea may have remained unreacted, contributing to the increase in the adhesive pH, since aqueous urea solutions are slightly alkaline (pH 8–9). A similar increase in the pH following the incorporation of kraft lignin into urea–formaldehyde adhesives has also been reported by Luckman et al. (2021) and Ferreira (2017) [1,52].

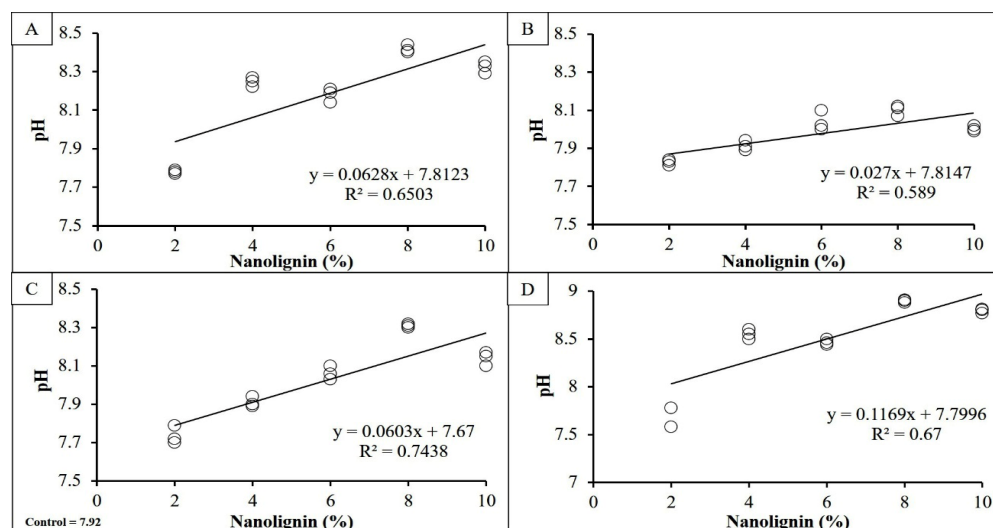


Figure 3. The mean pH values of the adhesives as a function of the lignin nanoparticle additions: (A) pine lignin nanoparticles: 5 passes; (B) pine lignin nanoparticles: 10 passes; (C) eucalyptus lignin nanoparticles: 5 passes; (D) eucalyptus lignin nanoparticles: 10 passes.

The greater increase in the pH observed for eucalyptus lignin nanoparticles produced after 10 milling passes may be attributed to their smaller particle size and higher specific surface area. These characteristics likely enhanced the exposure and accessibility of alkaline functional groups, resulting in a higher pH value.

As the lignin nanoparticle content increased, the viscosity of the adhesive formulations also increased (Figure 4). During adhesive synthesis, the acid stage plays a key role in determining the final viscosity. In this stage, condensation reactions promote the formation of methylene and methylene-ether linkages between urea molecules, producing urea–formaldehyde polymers with medium to high molecular weights and, consequently, increasing the adhesive viscosity [52,53].

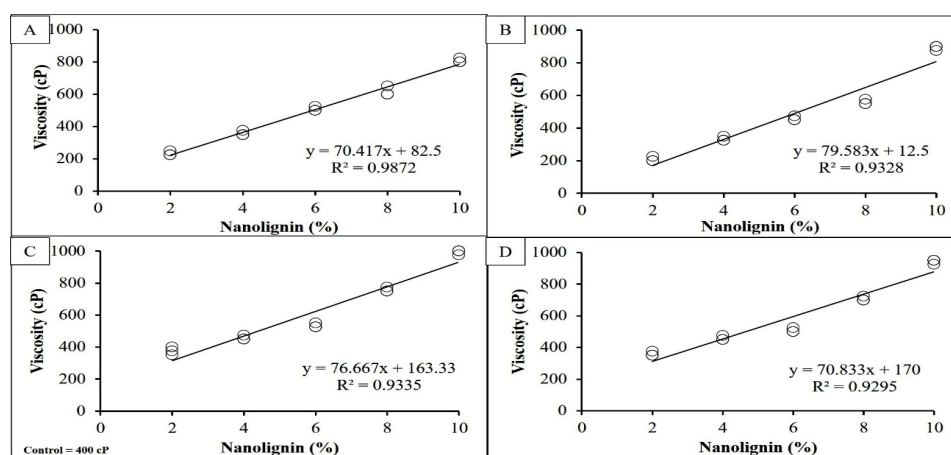


Figure 4. The mean viscosity values of modified urea–formaldehyde adhesives as a function of the lignin nanoparticle additions: (A) pine lignin nanoparticles: 5 passes; (B) pine lignin nanoparticles: 10 passes; (C) eucalyptus lignin nanoparticles: 5 passes; (D) eucalyptus lignin nanoparticles: 10 passes.

Among the evaluated treatments, the regression model for eucalyptus 10-pass lignin nanoparticles showed the highest intercept and steepest slope, indicating both a higher initial viscosity and a greater increase in viscosity with the increasing lignin nanoparticle content. This behavior is likely associated with the smaller particle size and higher specific surface area of these nanoparticles, which may increase the accessibility of reactive

functional groups during synthesis. Consequently, hydroxymethylation and subsequent condensation reactions may be enhanced, contributing to the formation of a more interconnected polymer network and, therefore, to the higher viscosity observed.

The duration of the acid stage was kept constant for all adhesive formulations. Therefore, formulations containing higher lignin nanoparticle contents likely underwent a greater extent of condensation during this stage, resulting in adhesives with higher viscosity. Adhesives formulated with eucalyptus lignin nanoparticles exhibited higher viscosity than those containing pine lignin nanoparticles. Eucalyptus lignin undergoes more extensive chemical degradation during the kraft pulping process because hardwood lignins contain a higher proportion of other linkages, which are more susceptible to cleavage under kraft conditions [54,55]. This degradation increases the availability of hydroxyl groups, which can form hydrogen bonds with neighboring molecules [56]. In addition, particle size also influences adhesive viscosity [57]. The smaller particle size and lower degree of agglomeration observed for eucalyptus lignin nanoparticles provide a higher specific surface area, favoring interactions with the urea–formaldehyde resin matrix. Therefore, the reacted eucalyptus lignin nanoparticles may have participated more effectively in the polymerization process, contributing to the higher viscosity of the adhesive formulations.

The solid contents of the adhesives were not affected by the treatments and did not differ significantly from that of the control formulation, with an average value of 50.02%. This result was expected because the added lignin nanoparticles replaced an equivalent mass of urea during adhesive synthesis. Consequently, the total solid contents remained comparable to that of the control adhesive, minimizing its influence on viscosity.

The dry shear strengths of wood joints bonded with the modified urea–formaldehyde adhesives are shown in Figure 5. Adhesives containing 4% eucalyptus lignin nanoparticles or 6% pine lignin nanoparticles, regardless of the number of milling passes, exhibited significantly higher shear strengths than the control adhesive. On average, these formulations increased the shear strength by approximately 10% relative to the control. As expected, the shear strength of solid pine wood (6.91 MPa) was higher than that of the bonded joints. The superior performance of the eucalyptus-based formulations is likely associated with the smaller particle size and more homogeneous morphology of the nanoparticles, which may have promoted better dispersion within the adhesive matrix and more effective interactions with the resin, resulting in improved bond strength.

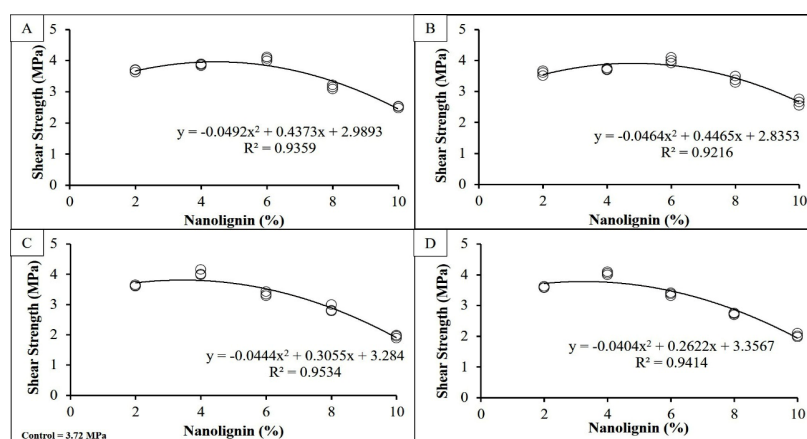


Figure 5. The mean shear strength (dry condition) values of the modified urea–formaldehyde adhesives as a function of the lignin nanoparticle additions: (A) pine lignin nanoparticles: 5 passes; (B) pine lignin nanoparticles: 10 passes; (C) eucalyptus lignin nanoparticles: 5 passes; (D) eucalyptus lignin nanoparticles: 10 passes.

Additions of 8 and 10% lignin nanoparticles resulted in significantly lower shear strength compared to the control adhesive. Chen et al. (2019) also reported that lignin nanoparticles can improve the shear strength of adhesives but emphasized that their concentration plays a critical role [26]. Because lignin nanoparticles are less reactive than urea, excessive incorporation may reduce the extent of crosslinking within the polymer network. Under these conditions, lignin nanoparticles tend to behave predominantly as fillers rather than as reactive components, resulting in lower bond strength.

The wet shear strengths of wood joints bonded with the modified urea–formaldehyde adhesives are shown in Figure 6. Adhesives containing 4 and 6% eucalyptus lignin nanoparticles and 6% pine lignin nanoparticles, regardless of the number of milling passes, exhibited wet shear strength values approximately 13% higher than that observed for the control adhesive (2.99 MPa). The improvement may be attributed to the incorporation of lignin nanoparticles into the urea–formaldehyde network. Their hydrophobic nature and reactive functional groups may reduce the susceptibility of the polymer network to hydrolytic degradation by interacting with hydroxymethyl groups and consuming residual free formaldehyde during curing. As a result, the degradation of methyl and methylene-ether linkages under wet conditions may be reduced, contributing to improve bond durability and high wet shear strength [27].

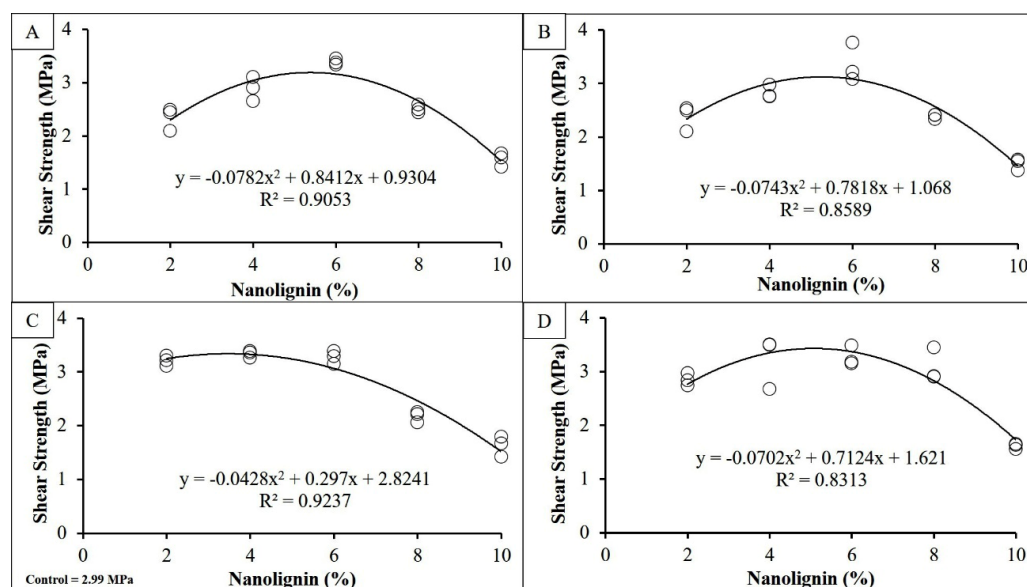


Figure 6. The mean shear strength (wet condition) values of the modified urea–formaldehyde adhesives as a function of the lignin nanoparticle additions: (A) pine lignin nanoparticles: 5 passes; (B) pine lignin nanoparticles: 10 passes; (C) eucalyptus lignin nanoparticles: 5 passes; (D) eucalyptus lignin nanoparticles: 10 passes.

However, the wet shear strength remained lower than the corresponding dry shear strength. This result highlights the need for further studies to improve the moisture resistance of these adhesives, particularly through optimization of the formaldehyde-to-urea molar ratio and adhesive formulation.

When the lignin nanoparticle content exceeded 6%, a significant reduction in the shear strength was observed under both dry and wet conditions. This behavior is likely associated with the substantial increase in the adhesive viscosity, which limits adhesive flow and mobility during bonding. Reduced fluidity restricts adhesive penetration into the wood structure, resulting in a thicker glue line with lower mechanical performance [58]. In addition, the morphology of the lignin nanoparticles and the greater tendency for particle agglomeration at higher loadings may further increase flow resistance and promote a less

homogeneous distribution of the adhesive within the wood substrate, thereby weakening interfacial bonding and reducing the shear strength.

3.3. FTIR and Quantification of Formaldehyde Emissions from Adhesives

FTIR analysis and formaldehyde emission measurements were performed only on the adhesives that exhibited the highest shear strengths and viscosity values of 400 cP or lower, as selected by the cluster analysis (Figure 7).

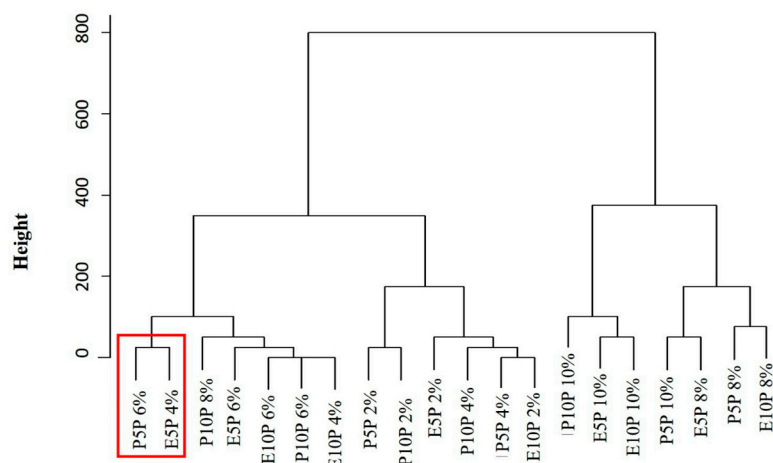


Figure 7. Clustering dendrogram. Adhesives containing P5P (pine lignin nanoparticles: 5 passes); P10P (pine lignin nanoparticles: 10 passes); E5P (eucalyptus lignin nanoparticles: 5 passes); and E10P (eucalyptus lignin nanoparticles: 10 passes) with percentages of 2, 4, 6, 8 and 10% lignin nanoparticles.

The similarity among treatments is represented by the distance between them in the dendrogram: the shorter the distance, the greater the similarity. Therefore, treatments containing 4% eucalyptus lignin nanoparticles and 6% pine lignin nanoparticles were selected for characterization, both produced after five milling passes.

The selected adhesive formulations were analyzed by infrared spectroscopy (FTIR) to evaluate possible chemical changes in their compositions resulting from the addition of lignin nanoparticles (Figure 8).

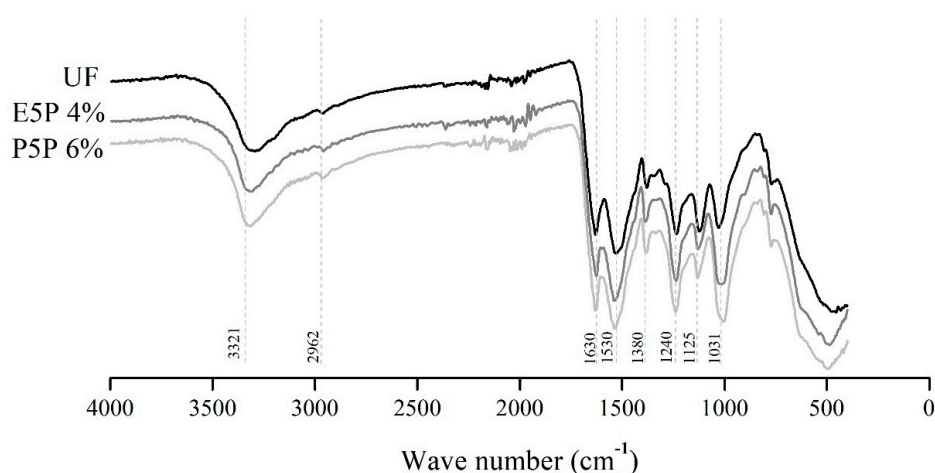


Figure 8. FTIR of adhesives. UF (urea–formaldehyde adhesive); E5P 4% (adhesives with 4% eucalyptus lignin nanoparticles—5 passes); P5P 6% (adhesives with 6% pine lignin nanoparticles—5 passes).

The broad absorption band at 3321 cm^{-1} is related to N–H stretching vibrations of aliphatic primary amines and O–H stretching of hydroxyl groups [55,56]. The weak band at 2962 cm^{-1} is attributed to symmetric C–H stretching vibrations of aliphatic ether, CH_2OH ,

and N-CH₂. The absorption band at 1630 cm⁻¹ is associated with stretching of the C=O bond of the primary amide.

The FTIR spectra of the modified urea–formaldehyde adhesives (E5P 4% and P5P 6%) exhibited subtle but relevant spectral changes that can be attributed to lignin nanoparticle incorporation, although many of these signals overlap with the characteristic bands of the matrix. A key lignin-related contribution can be associated with the region at 1530 cm⁻¹, where aromatic skeletal vibrations of the lignin structure typically occur (C=C stretching in the aromatic ring). In the modified adhesives, the slight changes in intensity and band shape in this region suggest the presence of lignin aromatic structures embedded within the UF network, partially overlapping with C-N vibrations of urea–formaldehyde [1].

The band at 1380 cm⁻¹ is assigned to the angular strain of CH₂ and CH₃. The bands at 1240 and 1031 cm⁻¹ correspond to the C-N stretching and angular deformation of NH₂ and the stretching of C-N bonds or N-C-N of methylene-ether bonds, respectively [59]. The band at 1240 cm⁻¹ can also be associated with C–O stretching in lignin [60]. Additionally, the region at 1030 cm⁻¹, associated with C–O stretching in secondary alcohols and ether linkages, may also reflect lignin contributions [61].

An increase in the intensity of these peaks was observed in the adhesives containing lignin nanoparticles, compared to the control adhesive. In addition, a small reduction in the 1125 cm⁻¹ band was observed, corresponding to C–O of aliphatic ether in the adhesives containing lignin nanoparticles [59]. Importantly, no new distinct peaks were observed, indicating that lignin incorporation did not introduce new covalent functional groups but rather contributed through overlapping vibrational modes and intermolecular interactions with the UF matrix.

From 180 °C, the temperature used for polymerization of the adhesives in this study, the degradation of the methylene-ether bonds of urea–formaldehyde adhesives begins, extending to temperatures close to 230 °C [62]. Therefore, the changes in intensity at peaks 1240 and 1031 cm⁻¹ may be related to the reaction of the lignin nanoparticles with the hydroxymethylureas formed and the possible connections with the terminal groups after the polymerization reaction, acting as blockers, and contributing to reducing or preventing the hydrolysis of urea with the formaldehyde.

Corroborating the results obtained in the FTIR analysis, a reduction in formaldehyde emissions was observed in the adhesives produced with lignin nanoparticles, compared to the control adhesive (Figure 9).

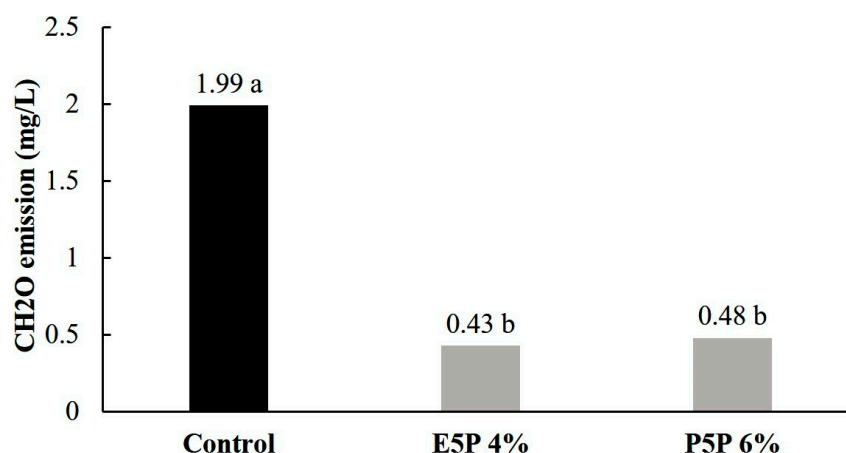


Figure 9. Emissions of formaldehyde by adhesives. Control (urea–formaldehyde adhesive); E5P 4% (adhesives with 4% eucalyptus lignin nanoparticles—5 passes); P5P 6% (adhesives with 6% pine lignin nanoparticles—5 passes). Different lowercase letters indicate statistically significant differences between treatments according to Tukey’s test ($p < 0.05$).

For the control adhesive (UF), the formaldehyde emission was 1.99 mg/L. For the E5P 4% and P5P 6% adhesives, the emissions were 0.43 and 0.48 mg/L, respectively, with an average emission reduction of around 75%. Both were classified as class E0 by the Chinese standard GB 18580 (2001), characterized by the lowest formaldehyde emission class, and as class F*** by the [63] standard JAS 233 (2003), which requires emissions ≤ 0.5 mg/L by the desiccator method [64,65].

The reductions in formaldehyde emissions achieved for adhesives produced with lignin nanoparticles may be associated with the lower hygroscopicity of lignin nanoparticles compared to urea, reducing the hydrolysis of existing bonds in the adhesives, and the greater number of crosslinks formed between their chains [63]. As a result, there is less release of the formaldehyde molecule during the formation of the three-dimensional network, which increases the mechanical shear strength of the glued joints, as verified.

In addition to this, the incorporation of lignin nanoparticles increased the adhesive viscosity, which may have reduced the mobility of the adhesive components during curing, favoring the retention of formaldehyde within the polymeric network. However, the higher viscosity can also impair adhesive flow and penetration into the wood substrate, limiting the formation of effective mechanical interlocking and interfacial interactions. Consequently, this effect may have contributed to the lower shear strength observed in the bonded joints containing lignin nanoparticles in high concentrations [18]. Thus, the reduction in formaldehyde emissions is likely associated with the combined effects of formaldehyde scavenging, physical diffusion barriers, and modifications in the adhesive microstructure promoted by lignin nanoparticle incorporation.

4. Conclusions

The additions of pine and eucalyptus kraft lignin nanoparticles into urea–formaldehyde adhesives promoted an increase in the pH and viscosity values. Among the evaluated treatments, the formulations containing 4% eucalyptus lignin nanoparticles (E5P 4%) and 6% pine lignin nanoparticles (P5P 6%), both produced after five milling passes, exhibited the best overall performances. These treatments increased the shear strength by up to 10% under dry conditions and by up to 13% under wet conditions compared with the control adhesive.

FTIR analysis suggested interactions between lignin nanoparticles and the hydroxymethylurea species formed during urea–formaldehyde polymerization. The modified adhesives exhibited increased intensification of the bands associated with C-N, N-C-N and NH₂ vibrations related to methylene-ether linkages, together with a slight decrease in the intensity of the band assigned to C-O aliphatic ether groups.

Reductions in formaldehyde emissions were up to 75% with the additions of E5P 4% and P5P 6% compared with the control adhesive. These results demonstrate that lignin nanoparticles can partially replace urea in the urea–formaldehyde adhesive formulations while improving the adhesive performance and substantially reducing formaldehyde emissions, thereby contributing to the development of more sustainable wood adhesives.

Overall, the results demonstrate that kraft lignin nanoparticles can be incorporated into urea–formaldehyde adhesives at appropriate concentrations to reduce formaldehyde emissions while maintaining or improving the bonding performance under the conditions evaluated in this study. However, these findings are limited to the experimental conditions evaluated in the present work. Therefore, further studies are recommended to investigate formaldehyde emissions over time under different environmental humidity conditions, curing kinetics and urea–formaldehyde–lignin interactions, as well as the quantification of free formaldehyde in liquid adhesives. Such investigations would contribute to a deeper

understanding of the mechanisms responsible for formaldehyde reduction and the long-term performance of lignin nanoparticle-modified urea–formaldehyde adhesives.

Author Contributions: T.A.L., A.d.C.O.C. and T.S.V. conducted the main investigation, data curation and writing—original draft preparation. G.H.D.T., I.F.D., M.R.C., V.O.d.P.B., A.M.M.L.C. and L.C.P. conducted the data curation and writing—original draft preparation. T.V.d.O., J.S., F.M. and M.J. conducted the writing—review and editing. A.d.C.O.C. and I.F.D. conducted the conceptualization, writing—review and editing, and project administration. All authors contributed to this review paper. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported by CAPES [Finance Code 001], and The APC was funded by the CISAS—Center for Research and Development in Agrifood Systems and Sustainability [UID/05937/2025 (<https://doi.org/10.54499/UID/05937/2025>)].

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding authors.

Acknowledgments: We are thankful for the funding from the Minas Gerais State Research Foundation (FAPEMIG), the Brazilian National Council for Science and Technology Development (CNPq), and Coordination for the Improvement of Higher Education Personnel (CAPES). We also thank the CISAS—Center for Research and Development in Agrifood Systems and Sustainability [UID/05937/2025 (<https://doi.org/10.54499/UID/05937/2025>)].

Conflicts of Interest: The authors declare no conflicts of interest.

References

1. Ferreira, J.C. Synthesis of Urea-Formaldehyde Adhesives with Addition of Kraft Lignin and Nanocrystalline Cellulose. Ph.D. Thesis, Federal University of Viçosa, Viçosa, Brazil, 2017.
2. Lengowski, E.C.; Bonfatti Júnior, E.A.; Nishidate Kumode, M.M.; Carneiro, M.E.; Satyanarayana, K.G. Nanocellulose-Reinforced Adhesives for Wood-Based Panels. In *Sustainable Polymer Composites and Nanocomposites*; Springer: Cham, Switzerland, 2019; pp. 1001–1025. [[CrossRef](#)]
3. Vital, B.R.; Maciel, A.D.S.; Della Lucia, R.M. Qualidade de Juntas Coladas Com Lâminas de Madeira Oriundas de Três Regiões Do Tronco de Eucalyptus Grandis, Eucalyptus Saligna e Pinus Elliottii. *Rev. Árvore* **2006**, *30*, 637–644. [[CrossRef](#)]
4. Wibowo, E.S.; Park, B.D. The Role of Acetone-Fractionated Kraft Lignin Molecular Structure on Surface Adhesion to Formaldehyde-Based Resins. *Int. J. Biol. Macromol.* **2023**, *225*, 1449–1461. [[CrossRef](#)] [[PubMed](#)]
5. Ridho, M.R.; Lubis, M.A.R.; Nawawi, D.S.; Fatriasari, W. Optimization of Areca Leaf Sheath Lignin nanoparticles Synthesis by a Mechanical Method for in Situ Modification of Ultra-Low Molar Ratio Urea-Formaldehyde Adhesives. *Int. J. Biol. Macromol.* **2024**, *271*, 132614. [[CrossRef](#)] [[PubMed](#)]
6. Wang, H.; Cao, M.; Li, T.; Yang, L.; Duan, Z.; Zhou, X.; Du, G. Characterization of the Low Molar Ratio Urea–Formaldehyde Resin with ¹³C NMR and ESI–MS: Negative Effects of the Post-Added Urea on the Urea–Formaldehyde Polymers. *Polymers* **2018**, *10*, 602. [[CrossRef](#)] [[PubMed](#)]
7. Khonakdar Dazmiri, M.; Valizadeh Kiamahalleh, M.; Dorieh, A.; Pizzi, A. Effect of the Initial F/U Molar Ratio in Urea-Formaldehyde Resins Synthesis and Its Influence on the Performance of Medium Density Fiberboard Bonded with Them. *Int. J. Adhes. Adhes.* **2019**, *95*, 102440. [[CrossRef](#)]
8. Demuner, I.F.; Borges Gomes, F.J.; Gomes, J.S.; Coura, M.R.; Borges, F.P.; Macedo Ladeira Carvalho, A.M.; Silva, C.M. Improving Kraft Pulp Mill Sustainability by Lignosulfonates Production from Processes Residues. *J. Clean. Prod.* **2021**, *317*, 128286. [[CrossRef](#)]
9. Nargotra, P.; Sharma, V.; Wang, H.M.D.; Shieh, C.J.; Liu, Y.C.; Kuo, C.H. Biocatalysis for Lignin Conversion and Valorization: Driving Sustainability in the Circular Economy. *Catalysts* **2025**, *15*, 91. [[CrossRef](#)]
10. Mukhopadhyay, D.; Chang, C.; Kulsreshtha, M.; Gupta, P. Bio-Separation of Value-Added Products from Kraft Lignin: A Promising Two-Stage Lignin Biorefinery via Microbial Electrochemical Technology. *Int. J. Biol. Macromol.* **2023**, *227*, 307–315. [[CrossRef](#)] [[PubMed](#)]
11. Wibowo, E.S.; Lubis, M.A.R.; Park, B.D. Simultaneous Improvement of Formaldehyde Emission and Adhesion of Medium-Density Fiberboard Bonded with Low-Molar Ratio Urea-Formaldehyde Resins Modified with Nanoclay. *J. Korean Wood Sci. Technol.* **2021**, *49*, 453–461. [[CrossRef](#)]
12. Correa-Guillen, E.; Henn, K.A.; Österberg, M.; Dessbesell, L. Lignin’s Role in the Beginning of the End of the Fossil Resources Era: A Panorama of Lignin Supply, Economic and Market Potential. *Curr. Opin. Green Sustain. Chem.* **2025**, *54*, 101038. [[CrossRef](#)]

13. Coura, M.R.; Demuner, A.J.; Demuner, I.F.; Blank, D.E.; Magalhães Firmino, M.J.; Borges Gomes, F.J.; Macedo Ladeira Carvalho, A.M.; Costa, M.M.; Henrique Dos Santos, M. Technical Kraft Lignin from Coffee Parchment. *Nord. Pulp Pap. Res. J.* **2023**, *38*, 229–241. [CrossRef]
14. Younesi-Kordkheili, H.; Pizzi, A. Lignin-Based Wood Adhesives: A Comparison between the Influence of Soda and Kraft Lignin. *Int. J. Adhes. Adhes.* **2023**, *121*, 103312. [CrossRef]
15. Käsäkoski, S.; Kasipandi, S.; Ohra-aho, T.; Wirtanen, T.; Lehtonen, J.; Alonso, D.M.; Vila, F.; Rautiainen, S. Depolymerisation of γ -Valerolactone Organosolv Lignins with Unsupported Molybdenum-Based Catalysts. *ChemSusChem* **2026**, *19*, e202500643. [CrossRef] [PubMed]
16. Zhao, W.; Simmons, B.; Singh, S.; Ragauskas, A.; Cheng, G. From Lignin Association to Nano-/Micro-Particle Preparation: Extracting Higher Value of Lignin. *Green Chem.* **2016**, *18*, 5693–5700. [CrossRef]
17. Kim, D.; Cheon, J.; Kim, J.; Hwang, D.; Hong, I.; Kwon, O.H.; Park, W.H.; Cho, D. Extraction and Characterization of Lignin from Black Liquor and Preparation of Biomass-Based Activated Carbon There-From. *Carbon Lett.* **2017**, *22*, 81–88. [CrossRef]
18. Pizzi, A.; Mittal, K.L. *Handbook of Sealant Technology*; CRC Press: Boca Raton, FL, USA, 2009; pp. 1–554. [CrossRef]
19. Viktoria, L.; Gottfried, A.; Pia, S.R.; Friedrich, S.; Christoph, G.; Jakob, N.; Hendrikus, v.H.; Paulik, C. Agricultural Residue Lignin from Novel Low-Temperature Pretreatment as Potential Raw Material for LPF Resins. *Biomass Convers. Biorefinery* **2023**, *14*, 17905–17912. [CrossRef]
20. Zhao, D.; Li, Z.; Zhang, Y.; Fu, P. The Role of Lignin in Adhesives for Lignin-Based Formaldehyde-Based Resins: A Review. *Biomass Convers. Biorefinery* **2025**, *15*, 16307–16341. [CrossRef]
21. Hussin, M.H.; Appaturi, J.N.; Poh, N.E.; Latif, N.H.A.; Brosse, N.; Ziegler-Devin, I.; Vahabi, H.; Syamani, F.A.; Fatriasari, W.; Solihat, N.N.; et al. A Recent Advancement on Preparation, Characterization and Application of Lignin nanoparticles. *Int. J. Biol. Macromol.* **2022**, *200*, 303–326. [CrossRef] [PubMed]
22. Moreira, W.M.; Moreira, P.V.V.; dos Santos, D.F.; Gimenes, M.L.; Vieira, M.G.A. Nanogreen Is the New Future: The Conversion of Lignin and Lignocellulosic Wastes into Nanomaterials. *Environ. Sci. Pollut. Res.* **2023**, *30*, 19564–19591. [CrossRef] [PubMed]
23. Fatriasari, W.; Sohail, A.; Ridho, M.R.; Madyaratri, E.W.; Gillela, S.; Yadav, S.M.; Hamidon, T.S.; Hussin, M.H. Nanolignin in Bioadhesives for Wood Composites. In *Nanomaterials Additives in Bioadhesives for Wood Composites*; Springer: Singapore, 2025; pp. 83–117. [CrossRef]
24. Behboudi, G.; Shayesteh, K.; Tavakkoli Yarak, M.; Ebrahimi, H.A.; Moradi, S. Optimized Synthesis of Lignin Sulfonate Nanoparticles by Solvent Shifting Method and Their Application for Adsorptive Removal of Dye Pollutant. *Chemosphere* **2021**, *285*, 131576. [CrossRef] [PubMed]
25. Melro, E.; Alves, L.; Antunes, F.E.; Medronho, B. A Brief Overview on Lignin Dissolution. *J. Mol. Liq.* **2018**, *265*, 578–584. [CrossRef]
26. Chen, Y.; Gong, X.; Yang, G.; Li, Q.; Zhou, N. Preparation and Characterization of a Lignin nanoparticles Phenol Formaldehyde Resin by Replacing Phenol Partially with Lignin Nanoparticles. *RSC Adv.* **2019**, *9*, 29255–29262. [CrossRef] [PubMed]
27. Fleischer, O.; Marutzky, R. Hydrolyse von Harnstoff-Formaldehyd-Harzen: Auflösung Des Spangefüges in Holzwerkstoffen Durch Hydrolytischen Abbau Der Leimfuge. *Holz Als Roh-Und Werkst.* **2000**, *58*, 295–300. [CrossRef]
28. IARC Publications Website—IARC Monographs on the Identification of Carcinogenic Hazards to Humans. Available online: <https://publications.iarc.who.int/Book-And-Report-Series/Iarc-Monographs-On-The-Identification-Of-Carcinogenic-Hazards-To-Humans> (accessed on 26 June 2026).
29. Aakko-Saksa, P.T.; Lehtoranta, K.; Kuittinen, N.; Järvinen, A.; Jalkanen, J.P.; Johnson, K.; Jung, H.; Ntziachristos, L.; Gagné, S.; Takahashi, C.; et al. Reduction in Greenhouse Gas and Other Emissions from Ship Engines: Current Trends and Future Options. *Prog. Energy Combust. Sci.* **2023**, *94*, 101055. [CrossRef]
30. Almeida, M.L.; Gonçalves, F.D.; Martins, J.M.; Magalhães, F.D.; Ramos, R.M.; Carvalho, L.H. Addressing Formaldehyde Emissions in Wood-Based Panels: Evaluation of Production Processes, Measurement Strategies, and Novel Solutions. *J. Adhes.* **2025**, *101*, 1252–1274. [CrossRef]
31. Chen, L.; Long, Y.; Huang, B.; Sun, C.; Kang, S.; Wang, Y. Technological Innovations Review in Reducing Formaldehyde Emissions through Adhesives and Formaldehyde Scavengers for Building Materials. *Environ. Res.* **2025**, *273*, 121242. [CrossRef] [PubMed]
32. Wang, Q.Q.; Zhu, J.Y.; Gleisner, R.; Kuster, T.A.; Baxa, U.; McNeil, S.E. Morphological Development of Cellulose Fibrils of a Bleached Eucalyptus Pulp by Mechanical Fibrillation. *Cellulose* **2012**, *19*, 1631–1643. [CrossRef]
33. Mishra, P.K.; Ekielski, A. A Simple Method to Synthesize Lignin Nanoparticles. *Colloids Interfaces* **2019**, *3*, 52. [CrossRef]
34. Lopes, M.S. Obtaining and Characterizing Lignins and Nanoparticles of Klason and Kraft Lignin, 2018. Available online: <https://hdl.handle.net/1884/55219> (accessed on 15 February 2025).
35. D1490-01; Test Method for Nonvolatile Content of Urea-Formaldehyde Resin Solutions. American Society for Testing and Materials (ASTM): West Conshohocken, PA, USA, 2013.
36. ASTM E70-07; Test Method for PH of Aqueous Solutions with the Glass Electrode. ASTM International: West Conshohocken, PA, USA, 2015.

37. ASTM D1084-97; Test Methods for Viscosity of Adhesives. ASTM International: West Conshohocken, PA, USA, 1997.
38. ASTM D2339-98; Standard Test Method for Strength Properties of Adhesives in Two-Ply Wood Construction in Shear by Tension Loading. American Society for Testing and Materials (ASTM): West Conshohocken, PA, USA, 2004. Available online: https://standards.iteh.ai/catalog/standards/astm/f4e48f97-d2bc-4d66-a5da-ce9d52e370d5/astm-d2339-98?reviews=true&srsId=AfmBOodxUWcrJTNX4ZuBh72tqz2omcBb-Yb1JYet-dkxZ_g3-mhIUz8 (accessed on 26 June 2026).
39. ABNT NBR ISO 12466; Madeira Compensada—Qualidade de Colagem. Associação Brasileira de Normas Técnicas (ABNT): West Conshohocken, PA, USA, 2006. Available online: <https://pt.scribd.com/document/778510344/ABNT-NBR-ISO-12466-Madeira-Compensada-Qualidade-de-Colagem> (accessed on 26 June 2026).
40. ASTM D5582-14; Test Method for Determining Formaldehyde Levels from Wood Products Using a Desiccator. American Society for Testing and Materials: West Conshohocken, PA, USA, 2014.
41. Yan, P.; Xu, Z.; Zhang, C.; Liu, X.; Xu, W.; Zhang, Z.C. Fractionation of Lignin from Eucalyptus Bark Using Amine-Sulfonate Functionalized Ionic Liquids. *Green Chem.* **2015**, *17*, 4913–4920. [CrossRef]
42. Tejado, A.; Peña, C.; Labidi, J.; Echeverria, J.M.; Mondragon, I. Physico-Chemical Characterization of Lignins from Different Sources for Use in Phenol–Formaldehyde Resin Synthesis. *Bioresour. Technol.* **2007**, *98*, 1655–1663. [CrossRef] [PubMed]
43. Latham, K.G.; Matsakas, L.; Figueira, J.; Rova, U.; Christakopoulos, P.; Jansson, S. Examination of How Variations in Lignin Properties from Kraft and Organosolv Extraction Influence the Physicochemical Characteristics of Hydrothermal Carbon. *J. Anal. Appl. Pyrolysis* **2021**, *155*, 105095. [CrossRef]
44. Wright, I.A. Field and Laboratory Methods for Grassland and Animal Production Research. *Grass Forage Sci.* **2002**, *57*, 189. [CrossRef]
45. Inwood, J.P.W. Sulfonation of Kraft Lignin to Water Soluble Value Added Products. Doctoral Dissertation, Lakehead University, Thunder Bay, ON, Canada, 2014.
46. Gonugunta, P. Synthesis and Characterization of Biobased Carbon Nanoparticles from Lignin. Doctoral Dissertation, University of Guelph, Guelph, ON, Canada, 2012.
47. Melro, E.; Filipe, A.; Sousa, D.; Valente, A.J.M.; Romano, A.; Antunes, F.E.; Medronho, B. Dissolution of Kraft Lignin in Alkaline Solutions. *Int. J. Biol. Macromol.* **2020**, *148*, 688–695. [CrossRef] [PubMed]
48. Watkins, D.; Nuruddin, M.; Hosur, M.; Tcherbi-Narteh, A.; Jeelani, S. Extraction and Characterization of Lignin from Different Biomass Resources. *J. Mater. Res. Technol.* **2015**, *4*, 26–32. [CrossRef]
49. Lisperguer, J.; Perez, P.; Urizar, S. Structure and Thermal Properties of Lignins: Characterization by Infrared Spectroscopy and Differential Scanning Calorimetry. *J. Chil. Chem. Soc.* **2009**, *54*, 460–463. [CrossRef]
50. Ait Benhamou, A.; Abid, L.; Calvez, I.; Cloutier, A.; Nejad, M.; Stevanovic, T.; Landry, V. Advances in Lignin Chemistry, Bonding Performance, and Formaldehyde Emission Reduction in Lignin-Based Urea-Formaldehyde Adhesives: A Review. *ChemSusChem* **2025**, *18*, e202500491. [CrossRef] [PubMed]
51. Nair, S.S.; Sharma, S.; Pu, Y.; Sun, Q.; Pan, S.; Zhu, J.Y.; Deng, Y.; Ragauskas, A.J. High Shear Homogenization of Lignin to Lignin nanoparticles and Thermal Stability of Lignin nanoparticles-Polyvinyl Alcohol Blends. *ChemSusChem* **2014**, *7*, 3513–3520. [CrossRef] [PubMed]
52. Stramosk Luckman, S.; Bayestorff da Cunha, A.; Rios, A.; Zanatta, P. Influência Da Incorporação de Lignina Kraft à Resina Ureia-Formaldeído Nas Propriedades Tecnológicas de Painéis Aglomerados Convencionais. *Sci. For.* **2021**, *49*, e3527. [CrossRef]
53. Gadhav, R.V.; Mahanwar, P.A.; Gadekar, P.T. Lignin-Polyurethane Based Biodegradable Foam. In *Methods in Lignin Chemistry*; Lin, S.Y., Dence, C.W., Eds.; Springer Series in Wood Science; Springer: Berlin/Heidelberg, Germany, 1992.
54. Development of Polyurethane Nanocomposite Adhesives Based on Castor Oil. Master's Thesis, Universidade Federal do Rio Grande do Sul, Porto Alegre, Brazil, 2009. Available online: <https://lume.ufrgs.br/handle/10183/23938> (accessed on 10 October 2024).
55. Chiang, V.L.; Funaoka, M.; Chiang, V.L.; Funaoka, M. The Difference between Guaiacyl and Guaiacyl-Syringyl Lignins in Their Responses to Kraft Delignification. *Holzforschung* **1990**, *44*, 309. [CrossRef]
56. Bonfatti Junior, E.A. Qualidade da Lignina Kraft em Função do Agente Protonador. Ph.D. Thesis, Federal University of Paraná, Curitiba, Paraná, Brazil, 2020. Available online: <https://hdl.handle.net/1884/69847> (accessed on 26 June 2026).
57. Sanghvi, M.R.; Tambare, O.H.; More, A.P. Performance of Various Fillers in Adhesives Applications: A Review. *Polym. Bull.* **2022**, *79*, 10491–10553. [CrossRef]
58. Graham Solomons, T.W.; Fryhle, C.B.; Snyder, S.A. *Organic Chemistry*, 12th ed.; John Wiley & Sons: Hoboken, NJ, USA, 2016.
59. Roumeli, E.; Papadopoulou, E.; Pavlidou, E.; Vourlias, G.; Bikiaris, D.; Paraskevopoulos, K.M.; Chrissafis, K. Synthesis, Characterization and Thermal Analysis of Urea–Formaldehyde/NanoSiO₂ Resins. *Thermochim. Acta* **2012**, *527*, 33–39. [CrossRef]
60. Müller, S.M.; Schaubeder, J.B.; Maier, J.; Schlemmer, W.; Spirk, S. Sustainable Photoresists from Kraft Lignin. *Monatshefte Für Chem.–Chem. Mon.* **2025**, *157*, 459–469. [CrossRef]
61. Bykov, I. Characterization of Natural and Technical Lignins Using FTIR Spectroscopy. Master's Thesis, Luleå University of Technology, Luleå, Sweden, 2008.

62. Zhang, J.; Wang, X.; Zhang, S.; Gao, Q.; Li, J. Effects of Melamine Addition Stage on the Performance and Curing Behavior of Melamine-Urea-Formaldehyde (MUF) Resin. *BioResources* **2013**, *8*, 5500–5514. [[CrossRef](#)]
63. JPIC/JAS Plywood Standards (Japan)—Plywood Supplier & Manufacturer. Available online: <https://www.plywood.cc/jpicjas-plywood-standards-japan/> (accessed on 26 June 2026).
64. Zorba, T.; Papadopoulou, E.; Hatjiissaak, A.; Paraskevopoulos, K.M.; Chrissafis, K. Urea-Formaldehyde Resins Characterized by Thermal Analysis and FTIR Method. *J. Therm. Anal. Calorim.* **2008**, *92*, 29–33. [[CrossRef](#)]
65. GB 18580-2001; Indoor Decorating and Refurbishing Materials—Limit of Formaldehyde Emission of Wood Based Panels and Finishing Products. The Standardization Administration of the People's Republic of China: Beijing, China, 2001.

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.