Research Article

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Preparation of highly water-resistant wood adhesives using ECH as a crosslinking agent

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Abstract: In this study, kraft lignin and epichlorohydrin (ECH) were used to prepare no-formaldehyde wood adhesives. The lignin was first treated by ball milling, then reacted with glyoxal to produce glyoxalated lignin under alkaline conditions, and then blended with ECH to prepare lignin-based formaldehyde-free adhesive. The influence of the content of ECH on the physicochemical properties of the adhesives was explored, and the possible synthesis mechanism of the ECH-modified glyoxalated lignin adhesives (glyoxalated kraft lignin-epoxy [GKLE]) was investigated. The results show that ECH was beneficial to improving the plywood shear strength and water resistance; the plywood prepared with GKLE-50 adhesive displays comparable water resistance as phenol–formaldehyde resins and its wet shear strength (type I) was 1.05 MPa, exceeding the Chinese National Standards GB/T 9846-2015. Scanning electron microscopy analysis showed that the increase of ECH content promoted the adhesive to penetrate the wood to form glue nails, improving the wet shear strength of the plywood. Chemical analysis indicated that glyoxalation was used to introduce hydroxyethyl groups into the ortho positions of the aromatic rings of lignin, and then the ring-opening reaction between glyoxalated lignin and ECH occurred forming ether bonds. Overall, lignin has displayed great potential in replacing formaldehyde-based adhesives for industrial applications.

Keywords: lignin, glyoxal, epichlorohydrin

1 Introduction

Formaldehyde-based adhesives (including urea–formaldehyde, melamine–urea–formaldehyde, and phenolic adhesives) are the most widely used wood adhesives due to their excellent bonding strength, low production cost, and fast curing (1,2). However, formaldehyde is a carcinogenic material, which is released into the environment during the production and use of pressed wood, endangering human health (3). At present, a strategy to reduce the emissions of formaldehyde is to replace part of the formaldehyde to participate in the reaction with strong reactive aldehydes such as glyoxal and furfural (4,5), so as to reduce the amount of formaldehyde in the resin preparation process. But, this strategy does not completely avoid the use of formaldehyde. The harmful effects of formaldehyde on human health have also prompted efforts to find formaldehyde-free adhesives alternatives. Bio-based adhesives, such as tannin, lignin, cellulose, and protein adhesives (6–8), are considered environmentally friendly and non-toxic adhesives that can be used as a substitute for formaldehyde-based adhesives (9).

Lignin is the most abundant polymer compound, second only to cellulose in the plant kingdom (10). As a by-product of the paper industry based on an alkaline delignification, kraft lignin is rarely used in chemical or material production but it is burned as energy for pulping mills (11). This underutilization stimulates the desire to develop lignin-based products of increased commercial value. The structure of lignin is similar to phenol, and the molecule contains active functional groups such as phenolic hydroxyl groups and aliphatic hydroxyl groups. Due to its unique structure, it can be used as a substitute for phenol to prepare phenolic resin (12,13). Although lignin has many functions, these potential active sites are often hidden or spatially obstructed by the high crosslinking density of lignin. Studies have shown that ball milling can increase the specific surface area of lignin, thereby improving the reaction activity (1). Except for physical methods to improve reactivity, traditional chemical modification methods are also effective, such as hydroxymethylation, glyoxylation (14), demethylation (15),

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and phenolization (16). Glyoxal is non-toxic and non-volatile (14,17), and it can be synthesized from renewable resources using existing technology, so this article chooses the glyoxylation method to modify lignin (18).

In this study, the industrial sulfate lignin was ball-milled first, and then the glyoxylation reaction was carried out. A combination of physical and chemical methods was used to increase the active sites of the lignin. Epichlorohydrin (ECH) as a crosslinking agent was used to prepare high water-resistant lignin-based formaldehyde-free wood adhesives (glyoxalated kraft lignin-epoxy [GKLE]). The influence of the addition of different amounts of ECH on the physical properties of GKLE adhesives was discussed. The wet shear strength of the samples was compared to evaluate the water resistance. The possible synthesis mechanism of lignin-based adhesives was studied by Fourier transform infrared spectroscopy (FTIR). The thermal stability of the adhesives was evaluated by differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). The microstructures of the bonded joints after the wet shear strength test of the plywood were observed by scanning electron microscopy (SEM).

2 Materials and methods

2.1 Materials

Industrial grade kraft lignin was obtained from J Hunan Taigreen Paper Group Hongjiang Paper Co., Ltd (Huaihua, China). The lignin was ground into particles of less than 120 μm by a ball mill and stored. Glyoxal (40 wt%, water solution) was purchased from Tianjin Bodi Chemical Co., Ltd (Tianjin, China). Sodium hydroxide was purchased from Chengdu Jinshan Chemical Reagent Co., Ltd (Chengdu, China). ECH was purchased from Tianjin Damao Chemical Reagent Factory. The chemicals used were of analytical grade and were used without further purification. Eucalyptus veneers were supplied by Qiaosheng Wood Industry Company in Guangxi province, Nanning, China.

2.2 Synthetic lignin-epoxy resin

Kraft lignin (125 g) was dissolved in 120 g of NaOH solution (30 wt%), and then glyoxal (90 wt% of the weight based on the total weight of lignin) was slowly added. The lignin solution was loaded into a 1 L round bottom flask equipped with a magnetic stirring rod and heated to 60°C. After stirring at 60°C for 4 h, the solution was cooled to room temperature to obtain glyoxalated kraft lignin (GKL). Then, the GKL solution and different contents of ECH (10%, 20%, 30%, 40%, and 50% based on the weight of GKL solution) were mixed mechanically to get a series of GKLE resins. The lignin-epoxy resins were named GKLE-10, GKLE-20, GKLE-30, GKLE-40, and GKLE-50, respectively, in which numbers represent the amount of ECH added.

2.3 Preparation of the three-layer plywood

To test the water resistance of the lignin-epoxy resins as plywood adhesives, we prepared three-ply plywood samples using the lignin-epoxy resin and eucalyptus veneer (200 mm × 200 mm × 1.8 mm). The layers were glued using adhesive with a glue spread rate of 150 $g{\cdot}m^{-2}$. The three-layer plywood was pre-pressed for 8 min at room temperature and then hot-pressed for 6 min pressure at 140°C under 8 MPa. The plywood was stored at room temperature for 24 h before testing.

2.4 Shear strength test

The dry and wet shear strengths of plywood were measured by a mechanical testing machine according to China National Standard GB/T 17657-2013 about test methods of evaluating the properties of wood-based panels. The plywood specimens were cut into 18 samples (size of 100 mm × 25 mm). A number of six specimens were randomly chosen for the dry shear strength test, and the others were for the wet shear strength test.

According to the requirement of type I and II of Chinese National Standard GB/T 17657-2013, two treatments were conducted for the wet shear strength test. For type II, the specimens were immersed in water at 63 ± 2°C for 3 h and then cooled to room temperature. For type I, the samples were immersed in boiling water for 4 h, then dried in the ventilated drying oven for 16–20 h at 60 ± 3°C, immersed again in boiling water for 4 h, and finally, cooled to room temperature before testing.

2.5 Characterization

The structural changes of samples were observed by FTIR (FTIR, Nicolet 6700 Nico-let Instrument Corporation, USA). The FTIR spectra were collected in the range of 4,000–400 cm^{-1} and at a resolution of 4 cm^{-1} by the KBr pellets method. The glass transition temperatures (T_g) of samples were measured

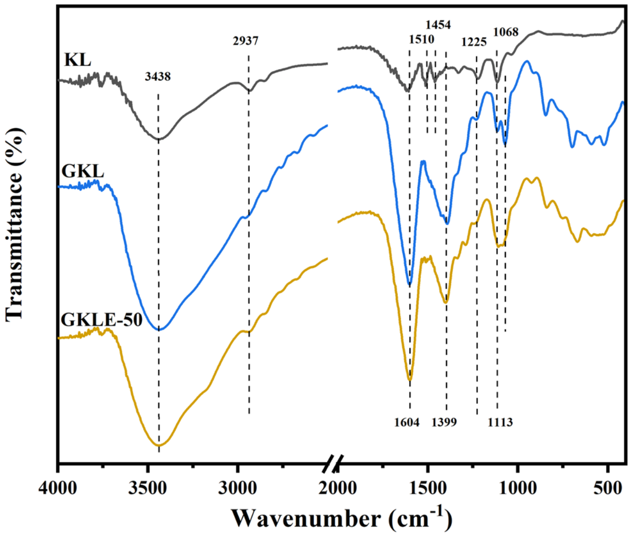


Figure 1: The FT-IR spectra of KL, GKL, and GKLE-50.

by DSC (DSC 214, Netzsch) from 40°C to 170°C with a heating rate of 10°C·min^{-1}. The thermal stability of KL, GKL, and fabricated GKLE-50 was determined by TGA (TGA, Q50, TA Instruments, Massachusetts, USA) with a rate of 10°C·min^{-1} in the presence of N_2, heating the samples from room temperature to 800°C. The microstructures of the bonded joints after the shear strength test were evaluated by SEM imaging on a Sigma 300 field emission electron microscope with an acceleration voltage of 3 kV. The SEM imaging magnifications were 200× and 2,000×.

3 Results and discussion

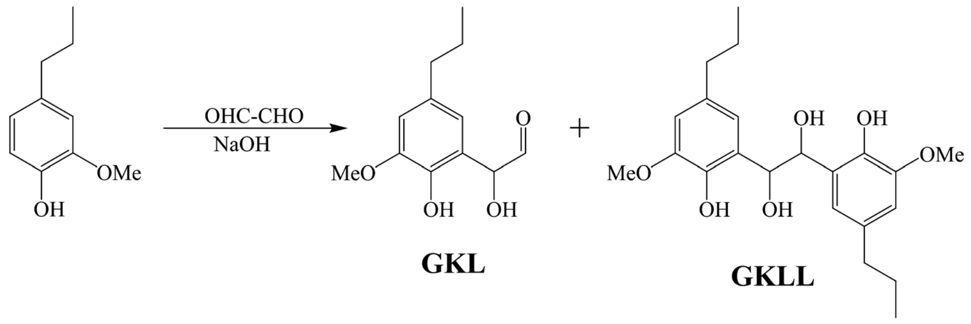
3.1 Synthesis mechanism of GKLE adhesives

FTIR analysis was used to understand the formation mechanism of GKLE adhesives. The FT-IR spectra of KL, GKL, and GKLE-50 are shown in Figure 1. A broad band at 3,438 cm^{-1} is visible in all of the samples, which corresponds to the O–H stretching vibrations in the phenolic and alcoholic structures of lignin. The peak at 2,937 cm^{-1} could be assigned to the C–H stretching vibration. The absorption peaks at 1,510, 1,454, and 1,399 cm^{-1} were attributed to the skeleton vibration of the aromatic ring. The peak at 1,604 cm^{-1} could be ascribed to the C═O stretching vibration of glyoxal. The absorption peak at 1,225 cm^{-1} was attributed to the stretching vibration of C–OH, and the absorption peaks at 1,113 and 1,068 cm^{-1} were attributed to the stretching vibration of C–O–C.

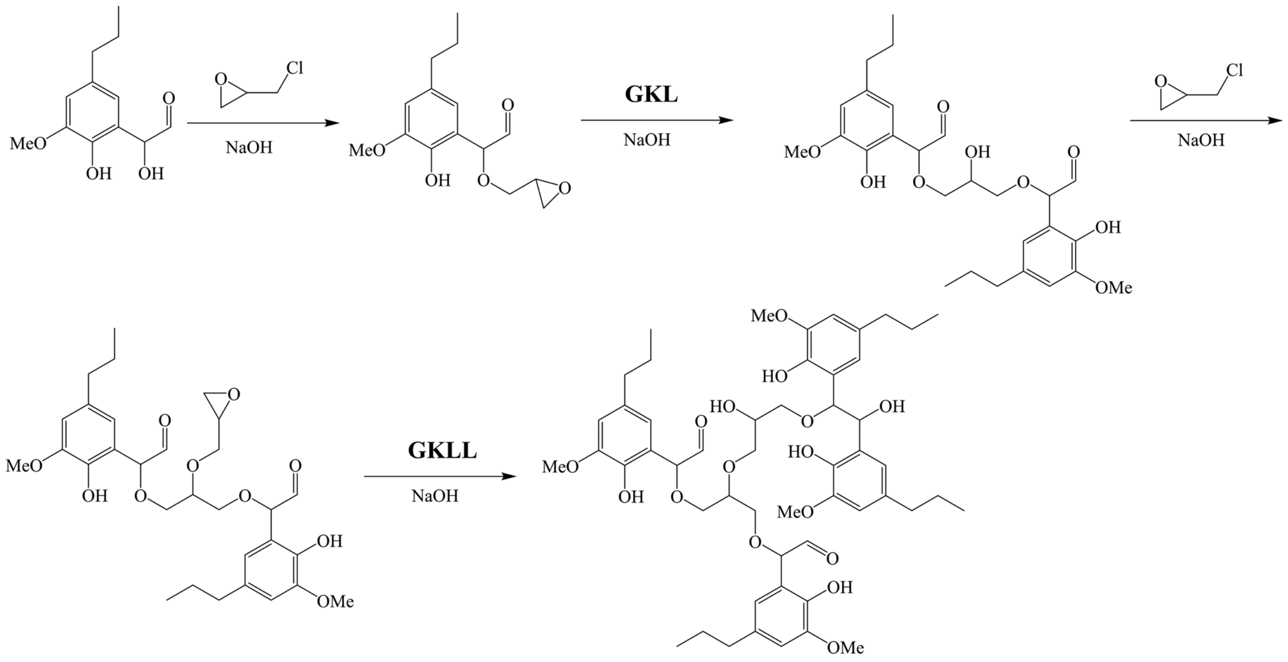
Compared with KL, the O–H signal of the GKL sample showed stronger O–H signals. Theoretically, the reaction of glyoxalation should increase the peak intensity of the O–H groups, indicating that there is formation of new hydroxyl groups among glyoxal and KL (Scheme 1) (19). In addition, the presence of C═O signals at 1,604 cm^{-1} again indicated that glyoxal did successfully react with KL. Compared to GKL, there is a reduction in the O–H band as seen in the GKLE-50 spectrum, indicating that the GKL was successfully cross-linked with ECH. The synthesis mechanism of LPFF adhesive is provided in Scheme 2.

3.2 DSC analysis

DSC analysis was used to investigate the effect of ECH content on the T_g temperature. The DSC curves and conversion degree curves of adhesives with different ECH contents are shown in Figures 2 and 3, respectively. The curing parameters of the adhesives were presented in Table 1 (ΔH is the enthalpy value). The DSC curve in Figure 2 shows that the curing reaction of the lignin-based formaldehyde-free adhesive is exothermic, and the enthalpy value GKLE-50 < GKLE-40 < GKLE-30 < GKLE-20 < GKLE-10. The result indicated that with the increase of



Scheme 1: Glyoxalation of lignin and glyoxal under alkaline conditions.



Scheme 2: Synthesis mechanism of LPFF adhesives.

ECH addition, the enthalpy value of the prepared lignin-based adhesive decreased gradually during the curing process, and the amount of heat emitted for the curing reaction decreased. The peak temperature is an important parameter to compare adhesive reactivity (20). It can be seen from Table 1 that the adhesive with a high ECH content showed higher reactivity, resulting in a shorter curing time of the adhesive (21). The results show that the curing temperature of the formaldehyde-free adhesive prepared with a high ECH content is also lower and can be cured at a lower temperature. As can be seen from the curing degree curve in Figure 3, the temperature of the adhesive first decreases and then increases with the increase of the amount of ECH when it reaches full curing, indicating that the curing of the adhesive first is easy and then becomes difficult. This may be because

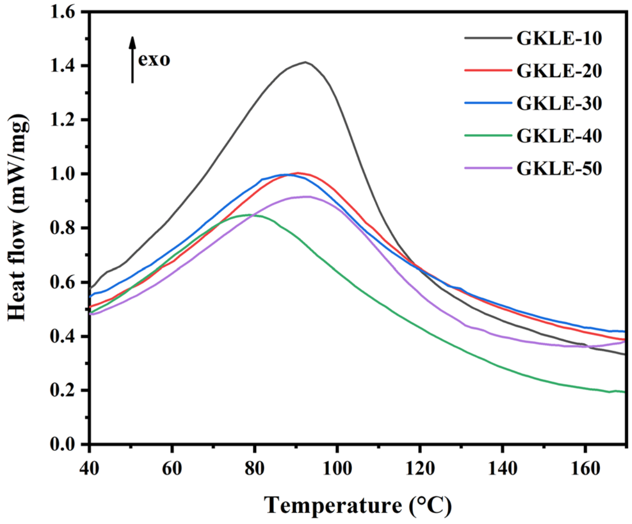


Figure 2: DSC curves of adhesives with different ECH contents.

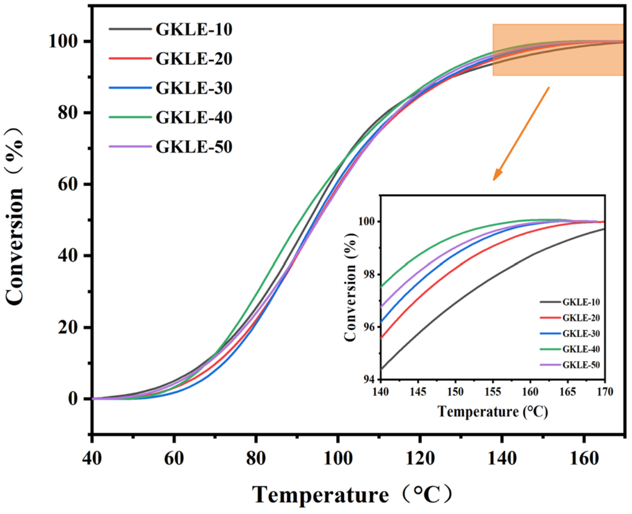


Figure 3: Conversion degree of adhesives with different ECH contents.

Table 1: Curing parameters of lignin-based adhesives

Adhesive	Δ*H* (J·g^{-1})	Initial temperature (°C)	End temperature (°C)	Peak temperature (°C)
GKLE-10	261	53	119	93
GKLE-20	173	51	122	91
GKLE-30	157	78	152	89
GKLE-40	138	47	169	83
GKLE-50	132	46	149	93

excessive ECH was not conducive to the ring-opening reaction.

3.3 Thermal stability analysis

TGA is usually used to evaluate the thermal stability of materials. In this study, the thermal stability and degradation of KL, GKL, and GKLE-50 were studied by TGA. The TGA and derivative thermogravimetry (DTG) curves of KL, GKL, and GKLE-50 are shown in Figure 4. As seen from Figure 4, the thermal degradation of KL, GKL, and GKLE-50 is similar, presenting three-stage decompositions. The thermal stability of GKL and GKLE-50 has been improved in contrast to KL. The initial weight loss of sulfated lignin, glyoxylated lignin, and adhesive occurs in the temperature range 40–150°C due

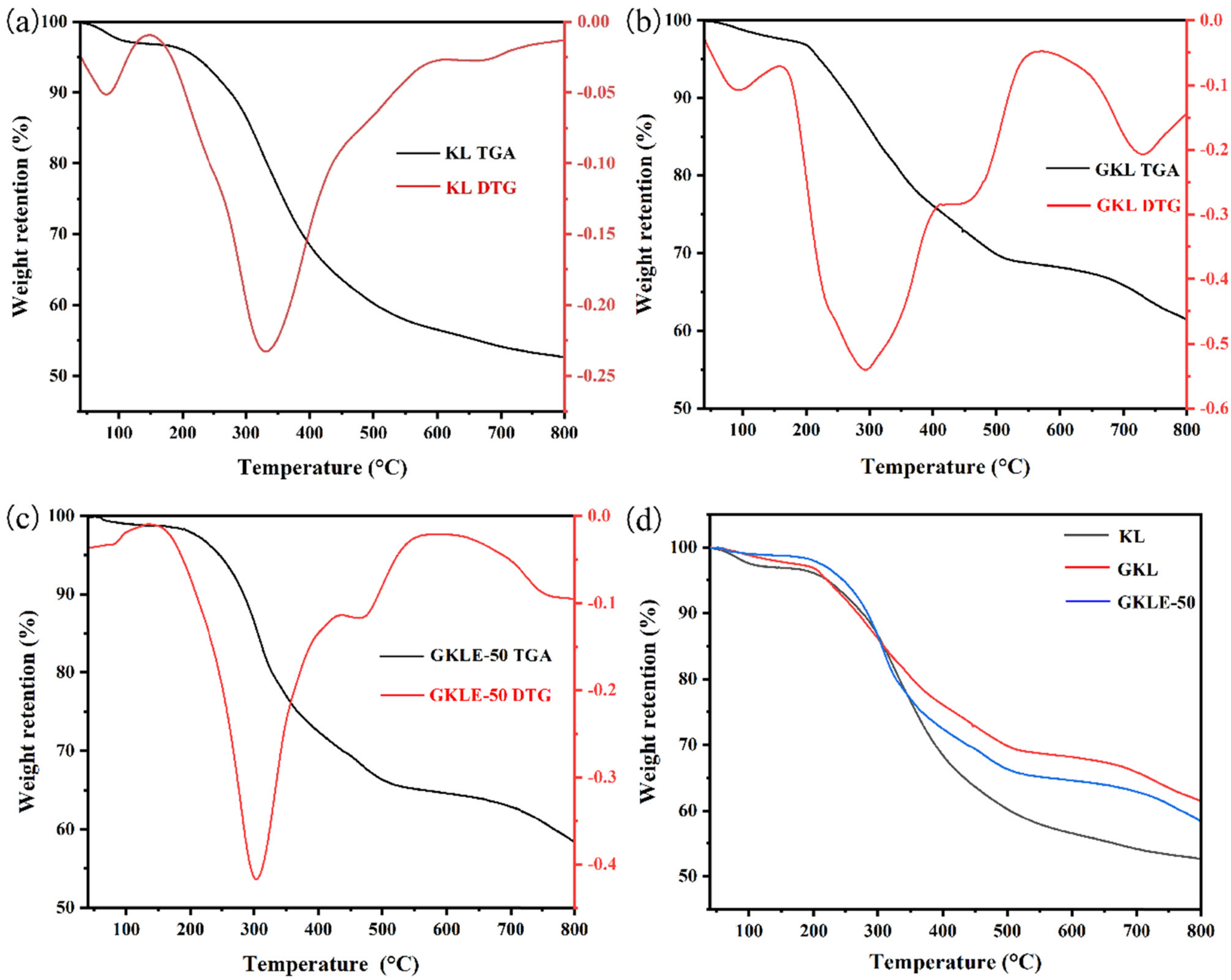


Figure 4: (a) TGA and DTG of KL; (b) TGA and DTG of GKL; (c) TGA and DTG of GKLE-50; (d) TGA of KL, GKL and GKLE-50.

Table 2: Results from thermal analysis for DTG_{max}, maximum decomposition rate, and residue

Sample	DTG_{max} (°C)	Maximum decomposition rate (%·min^{-1})	Residue (%)
KL	331	0.2	53
GKL	294	0.5	61
GKLE-50	303	0.4	58

to the evaporation of water molecules and the volatilization of small molecules. In the second stage, the main weight loss occurs in the temperature range 150–560°C, which is attributed to the breaking of ether bonds, methylene, and methine bridges in the molecular chain. After 560°C, the weight loss is due to further degradation of the polymer and volatilization of small molecules. As shown in Figure 4 and Table 2, the residual mass percentages of KL, GKL, and GKLE-50 at 800°C were 52.71%, 61.45%, and 58.42%, respectively. The result indicated that GKL has better thermal stability than that of KL due to glyoxylation increasing the molecular weight of sulfate lignin; meanwhile, the thermal stability of GKL is also better than that of GKLE-50 due to the poor thermal stability of ECH.

3.4 Analysis of the wet shear surface morphology of plywood

The microstructures of the bonded joints after the wet shear strength test of plywood are represented in Figure 5. For GKLE-10 and GKLE-20, no wood fiber was observed on the bonded joints and there was an adhesive coating on the fractured surface. In addition, no adhesive was observed on the wood catheter and wood rays. The results indicated that the adhesive did not permeate into the wood and the glue

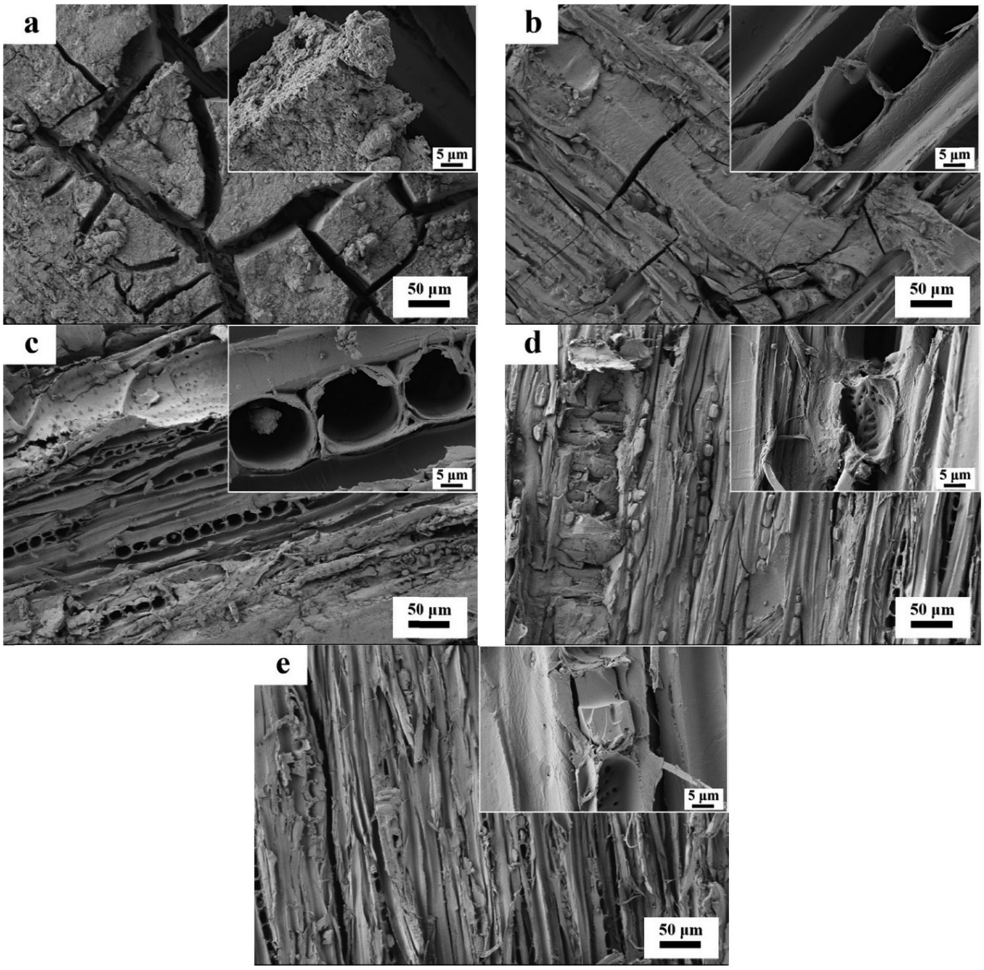


Figure 5: SEM images of bonded joints after the shear strength test of plywood: (a) GKLE-10, (b) GKLE-20, (c) GKLE-30, (d) GKLE-40, and (e) GKLE-50.

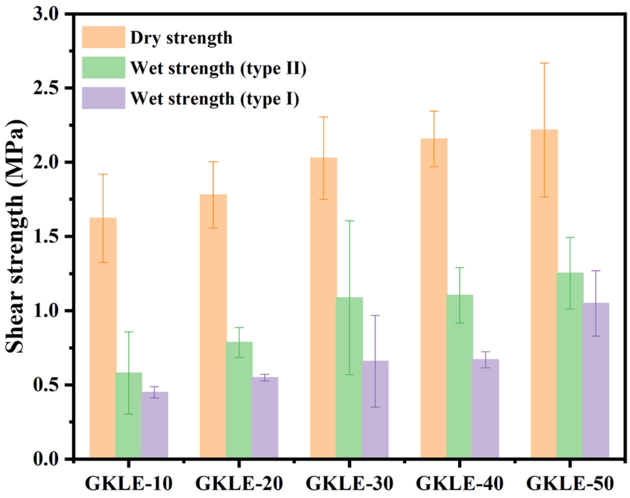


Figure 6: Effect of different ECH contents on the shear strength.

layer is brittly fractured under the action of an external force, causing poor mechanical performance (22,23). In contrast to GKLE-10 and GKLE-20, there are many tearing wood fibers on the wood surfaces of GKLE-30, GKLE-40, and GKLE-50. Furthermore, it was observed that the adhesive adhered to the wood pipes and wood rays to form glue nails. These results suggest that the increase of the ECH content promoted the adhesive to penetrate the wood to form glue nails, which improve the wet shear strength of the plywood. The higher the ECH content, the larger the glue nails and the greater the bonding quality of the adhesive. These results were consistent with the increase in the shear strength in the water-resistance test and the percentage of wood failure.

3.5 Water resistance of the fabricated plywood

The shear strengths by tension loading were used to study the adhesion performance of plywood, and it can be used to estimate the water resistance of the adhesive (23). The results of the dry and wet shear strength test on lignin-epoxy resins with different ECH contents are shown in Figure 6. The results indicated that ECH was beneficial to improving plywood shear strength and water resistance. For GKLE-10, the dry shear strength, wet shear strength (type II), and wet shear strength (type I) were 1.62, 0.58,

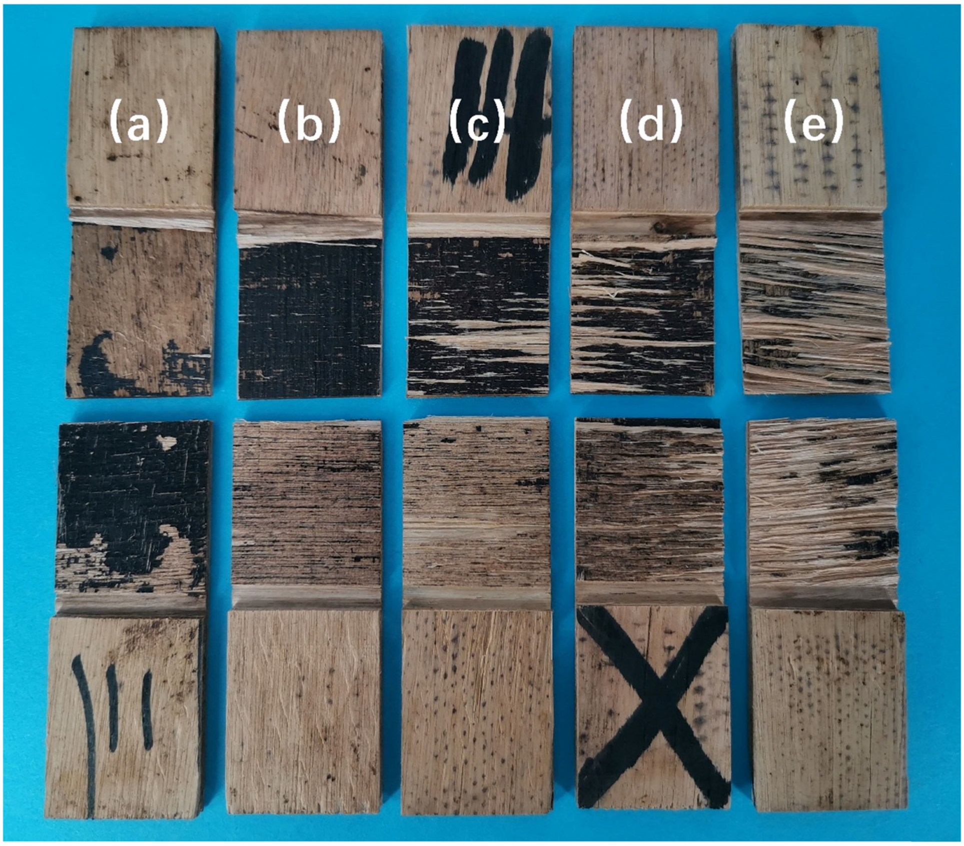


Figure 7: Influences of ECH on the wood failure of the wet plywood: (a) GKLE-10, (b) GKLE-20, (c) GKLE-30, (d) GKLE-40, and (e) GKLE-50.

and 0.45 MPa, respectively. In contrast to GKLE-10, the dry shear strength, wet shear strength (type II), and wet shear strength (type I) of GKLE-50 achieved an increase of 36.71%, 115.74%, and 133.33%, respectively. This result could be explained as follows: the addition of ECH improved the crosslinking density of the adhesive. A complex network structure was formed between ECH and GKL by a ring-opening reaction. In addition, the chemical reaction between the ether bond formed by the ring-opening reaction and the hydroxyl group in the wood cellulose could further improve the bonding strength of the plywood.

As shown in Figure 1, compared to the dry shear strengths of all plywoods, the wet shear strengths decreased significantly after one hot water (type II) immersion or two boiling water treatments (type II). This may be due to the swelling of the wood cellulose during treatment, which ultimately reduces the shear strength of the plywood (24). When the addition of ECH was 20–50%, the wet shear strengths (type II) of the plywood ranges from 0.79 to 1.25 MPa, satisfying the value (≥0.7 MPa) outlined in type II of Chinese standard GB/T 9846-2015. Thus, they were expected to replace the urea–formaldehyde resin (25). Furthermore, the wet shear strength (type I) of GKLE-50 was 1.05 MPa, which is higher than that specified for type I in the Chinese National Standard GB/T 9846-2015. Moreover, the water resistance and wet shear strength of the GKLE-50 resin are comparable to that of the phenolic resin.

The quality of the wood adhesive is usually determined by its shear strength and the percentage of wood failure (26). The wood failure percentage is high on behalf of great bonding quality. From Figure 7, it is observed that as the ECH content in the adhesive increases, the wood failure percentage increases. A lot of wood fiber tearing can be significantly observed in the plywood prepared by the GKLE-50 adhesive, indicating that the degree of wood fiber tearing becomes more obvious with the increase of ECH. This is because the epoxy-ring opening reaction between ECH and GKL, as well as the newly formed hydrogen bond between the C–O bond and the O–H bond on the wood enhances the interaction between the adhesive and the wood.

4 Conclusion

In this study, the reactivity of kraft lignin was improved by ball milling and glyoxalation. A formaldehyde-free lignin-based adhesive with good water resistance was prepared and ECH was used as a crosslinking agent. The physical and mechanical properties and the wood breaking rate of plywood analysis show that the water resistance and the wet shear strength of GKLE-50 were equivalent to those of the phenolic resin. Through FT-IR analysis, the possible mechanism of the formation of the adhesive polymerization reaction was speculated. Glyoxalated lignin undergoes a ring-opening reaction between the hydroxyl and ECH to form ether bonds, resulting in a dense network structure. DSC analysis indicated that the adhesive with a high ECH content has stronger reactivity and shorter curing time. DTG and TGA analysis showed that the thermal stability of glyoxalated lignin was improved due to the increase of molecular weight. The thermal stability of lignin-based adhesives decreased due to the addition of ECH but it was still higher than that of phenolic resin. In addition, SEM analysis showed that the increase in the ECH content promoted the penetration of the adhesive into the wood to form glue nails and improved the wet shear strength of the plywood. In summary, the excellent performance of lignin-based adhesives demonstrates the great potential of lignin in replacing formaldehyde-based adhesives, and at the same time facilitates the efficient development and utilization of biological resources.

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Data availability statement: All relevant data are presented in the manuscript file.

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