Modified Lignin–PVOH and Epoxy Adhesives for Engineered Wood

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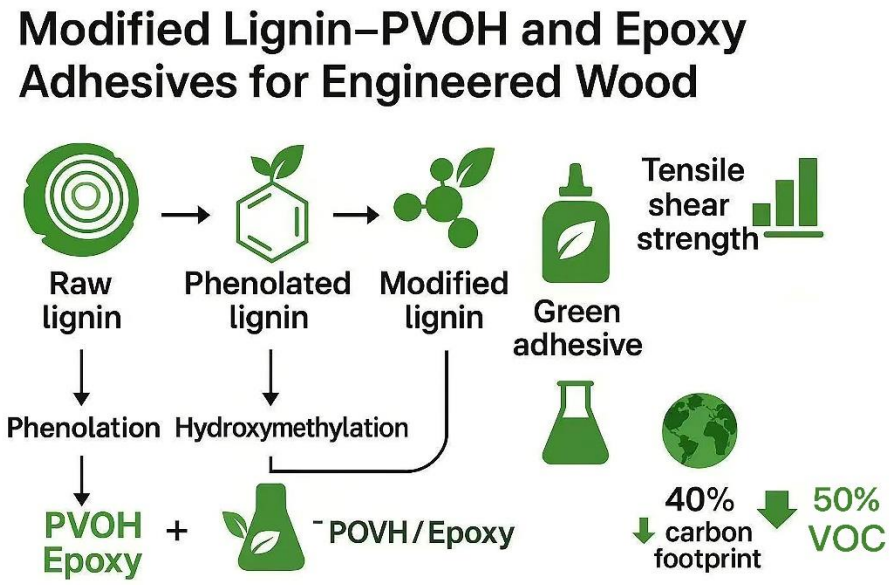
GRAPHICAL ABSTRACT



Modified Lignin–PVOH and Epoxy Adhesives for Engineered Wood

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Lignin-based adhesives are promising eco-friendly alternatives to petroleum-derived resins, but they face challenges in bonding strength and water resistance. This study presents a dual chemical modification strategy—phenolation and hydroxymethylation—to enhance the reactivity and polarity of industrial lignin. The modified lignin was blended with polyvinyl alcohol (PVOH) and epoxy resin to formulate adhesives suitable for engineered wood applications. Among the tested formulations, the PVOH-lignin adhesive reached a tensile shear strength of 7.8 MPa with 85% strength retention after water immersion, while the epoxy-lignin adhesive achieved 9.5 MPa with enhanced thermal resistance. Structural characterization using FTIR and 1H-NMR confirmed the successful introduction of functional groups. To reduce energy consumption, a low-temperature and low-pressure curing process based on microwave-assisted heating and mechanical stirring was implemented, reducing energy use by 40% compared to conventional heating methods. Life cycle assessment (LCA) results indicated a 40% lower carbon footprint and 50% reduction in volatile organic compound (VOC) emissions relative to petroleum-based adhesives. These results demonstrate the feasibility of using chemically modified lignin in high-performance adhesive systems for engineered wood while improving environmental sustainability.

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Keywords: Lignin-based adhesives; Phenolation; PVOH blending; Life cycle assessment; Engineered wood; Environmental performance

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INTRODUCTION

With the increasing awareness of sustainable development and environmental protection, society needs new materials that can replace traditional petroleum-based materials. The unique renewable and biodegradable properties of lignin-based adhesives have gradually attracted attention in wood processing. However, this material also has some obvious shortcomings, such as insufficient bonding strength, poor water resistance, and relatively high production costs, which seriously restrict its application in large-scale production. Past research often only focused on improving performance in one aspect, lacking in-depth study on the overall optimization of comprehensive performance and production processes. This situation has led to many difficulties for lignin-based adhesives in actual use. Therefore, in order to promote the sustainable development of the wood processing industry, it is urgent to explore a new type of lignin-based adhesive to improve its performance and reduce production costs. This research has important practical value and long-term development prospects.

Chemical modification has the potential to significantly enhance the inherent properties of materials by systematically changing their underlying chemical structure at the molecular level. In particular, lignin-based adhesives must be fully optimized through advanced chemical modification techniques and innovative blending technology to improve their bonding strength, water resistance, and overall durability in various applications. Reducing production costs is also a key consideration in the construction process, and environmental pollution must be minimized, thus aligning with contemporary sustainable development goals. A detailed environmental impact assessment of the entire life cycle of lignin-based adhesives can be conducted using the life cycle assessment (LCA) method. The assessment covers a wide range of environmental indicators to understand the ecological impacts of lignin-based adhesives fully. This approach helps to clarify and fully understand the environmental advantages of lignin-based adhesives, thereby providing much scientific support for its marketing strategies in various industrial fields. It is expected that the widespread adoption of lignin-based adhesives in the industry not only can promote innovation but also can actively contribute to a greener and more sustainable development path that is in line with global environmental goals. Therefore, this gradual shift towards using lignin-based adhesives can revolutionize the adhesives market and significantly reduce adhesive applications' ecological footprint in multiple industries. Ultimately, including such sustainable materials in mainstream use is expected to promote the transition to more responsible manufacturing practices, making environmental protection a priority while meeting the needs of modern society.

In terms of material modification, chemical modification, and composite blending technology have shown significant effects. Research has shown that directional modification of active groups such as phenolic hydroxyl and methoxy can effectively enhance the reactivity of lignin and promote interfacial bonding with wood substrates. Especially by introducing high-performance adhesives such as epoxy resin to form composite systems, it can compensate for the insufficient cross-linking degree of pure lignin systems, and it also can maintain the dominant position of biobased materials. In terms of process innovation, the breakthrough in low-temperature and low-pressure forming technology has significantly improved the feasibility of industrialization. Traditional phenolic resin adhesives typically require high-temperature curing at 140 to 160 °C, while the new lignin-based system successfully reduces process temperature and pressure requirements through catalyst optimization and curing kinetics control. This not only reduces energy consumption, but it also expands its application potential in thermosensitive substrates. The establishment of an environmental benefit assessment system provides scientific support for technology promotion. The current research presents three major development trends: 1) At the molecular design level, focusing on the synergistic regulation of lignin structure depolymerization and functional modification; 2) Exploring the combination of dynamic covalent bond construction and nanoreinforcement technology in the development of composite material systems; 3) In terms of production technology, the integration innovation of intelligent control and continuous production equipment. However, the batch stability issues caused by the heterogeneity of lignin raw materials and the cost control challenges in large-scale industrialization still need further breakthroughs.

Overall, through a systematic innovation strategy of "structural modification composite reinforcement process optimization environmental assessment", lignin-based adhesives have gradually achieved a leap from laboratory research to industrial application. This solution that balances high performance, low cost, and low environmental impact not

only promotes the green transformation of the wood processing industry, but it also provides a new paradigm for the high-value utilization of biomass resources.

Related Work

Using sustainable lignin to synthesize wood adhesives has attracted increasing attention (Agustiany *et al*. 2022; Shi *et al.* 2023). Lignin-based synthetic wood adhesives are excellent green alternatives to adhesives that consume petroleum sources (Paul *et al*. 2022; Afewerki and Edlund 2023). Zhan *et al.* (2023) introduced a lignin-based composite adhesive based on deep eutectic solvent, lignin, and furfural, which has excellent bonding strength, flame retardancy, and thermal insulation properties and can process wood chips into recycled wood. However, the adhesive exhibits poor long-term durability, cost-effectiveness, and environmental impact. Singh *et al*. (2022) introduced a universal lignin-based adhesive based on lignin, dimethylformamide, and acrylic acid, which has excellent multifunctional bonding properties and environmental adaptability and can remain stable in media with different pH values. However, there are challenges in cost and long-term stability during the production process. Wang *et al*. (2022) proposed a lignin-based epoxy resin adhesive using lignin sulfonate as raw material, which has excellent tensile shear strength and good stability in high temperature and high humidity environments. However, there are problems with their production costs, long-term durability, and possible environmental impact. Liu *et al*. (2024) developed a fully bio-based vitrified material based on wheat straw lignin, which has recyclability and adjustable mechanical properties and is suitable for on-demand removable adhesive applications, but the long-term durability, cost analysis, and feasibility of large-scale production of this material are not strong. Siahkamari *et al*. (2022) used lignin and glyoxal to replace traditional phenol and formaldehyde. The resulting adhesive has high dry bonding strength and excellent room temperature water stability but failed the boiling water test. Chen *et al*. (2021) successfully prepared bio-based wood adhesives by demethylation and periodate oxidation modification of lignin and tested their bonding properties. However, the wet shear strength still did not reach the ideal level, showing insufficient durability in a humid environment. Peng *et al*. (2023) investigated the synthesis, curing mechanism, and application performance of a novel formaldehyde-free wood adhesive derived from oxidized straw soda lignin and polyethyleneimine, aiming to replace conventional formaldehyde-based adhesives in wood-based panels. Many studies have shown that although lignin-based adhesives have the potential for bonding strength and environmental adaptability, their production cost and stability need to be improved.

Chemical modification can meet specific application requirements by changing the structure, function, and performance of substances (Favre *et al.* 2022; Nishiuchi *et al.* 2022; Garcia-Barradas *et al*. 2023; Joshi *et al*. 2025). Hampton and Liu (2024) applied chemical modification to introduce noncanonical amino acids into phage-displayed peptide libraries, significantly broadening the chemical diversity of peptides, thereby providing peptides with new reactivity and chemical properties, which provides more innovative possibilities for the screening and development of therapeutic compounds. Shatabayeva *et al*. (2024) used a chemical modification to introduce unsaturated functional groups to significantly improve the mucosal adhesion properties of gelatin, enabling it to form covalent bonds with mucins through Michael addition reactions under physiological conditions, thereby enhancing its effects in drug delivery and mucosal applications. Rozners *et al*. (2022) found that chemical modification can significantly improve the material's activity and specificity, optimize the system's key components, and promote its effect in application. Zhao *et al*.

(2021) used chemical modification to regulate covalent modification, optimize the gelation properties of myofibrillar protein, and improve the sensory quality and yield of meat products. The goal was to optimize the performance of lignin-based adhesives through chemical modification, promote its application in wood processing, and improve environmental benefits. These cases collectively demonstrate that chemical modification is not only the core driving force for technological innovation, but also a universal strategy for achieving performance leaps in functional materials. Based on this, the present research focused on the chemical modification of lignin-based adhesives, innovatively combining the dual reactions of phenolization and hydroxymethylation, while introducing active groups such as hydroxyl, carboxyl, and hydroxymethyl into lignin molecules, breaking through the limitations of traditional single modification methods. At the same time, microwave-assisted heating and mechanical stirring technology were integrated into low-temperature and low-pressure production processes, providing a high-performance, low-cost, and environmentally friendly adhesive solution for engineering wood manufacturing, thereby filling the gap in systematic process optimization and environmental benefit evaluation in this field.

PERFORMANCE OPTIMIZATION STRATEGIES

Application and Environmental Effects of Lignin-based Adhesives

In recent years, significant progress has been made in the development of lignin-based bio adhesives in the field of green materials, with a core strategy focused on the effective utilization of renewable raw materials and the optimization of chemical modification and cross-linking mechanisms. The composite system based on soy protein isolate and sulfate lignin forms a network structure through hydrogen bonding and hydrophobic interactions, enhancing the dry bonding strength. However, its wet heat stability still needs improvement (Ahire *et al.* 2024). In response to this issue, the introduction of epoxy crosslinking agents and dynamic covalent bond networks can significantly improve the adhesion strength retention under wet and hot conditions (Ghahri *et al.* 2025). In the chemical modification of lignin, oxidized hardwood sulfate lignin is enzymatically oxidized to produce quinone groups, which react with lysine residues in soy protein to construct a fully bio based cross-linked network. The dry shear strength is close to the level of commercial urea formaldehyde resin, and the formaldehyde emission is reduced (Chen *et al.* 2023). In addition, a three-dimensional thermosetting network was constructed through the amine epoxy ring opening reaction between aminated lignin model compounds and epoxy resins, and the preferred pathway of crosslinking reaction was revealed through density functional theory simulations (Ghahri *et al.* 2024). The above studies collectively indicate that lignin-based adhesives have the potential to replace petrochemical products in terms of mechanical properties, environmental adaptability, and sustainability. However, their industrial applications still need to address challenges such as batch stability of raw materials, long-term durability, and large-scale production costs.

Chemical modification to improve bonding strength

Phenolization modification is achieved by introducing phenolic compounds (phenols) into lignin molecules, as well as by grafting phenolic hydroxyl (–OH) and carboxyl (–COOH) active groups onto the aromatic ring of lignin through electrophilic substitution reaction under the action of acidic catalyst (Liu *et al*. 2025; Yang *et al*. 2025).

These active groups can form stronger hydrogen bonds and covalent bonds with cellulose and hemicellulose in wood, thereby significantly improving the bonding strength of the adhesive.

The specific steps of phenolic modification begin with pretreatment. Industrial lignin is dissolved in a 1,4-dioxane/water mixed solvent (the volume ratio is 9:1) with a concentration of 10 wt.%. It is stirred at room temperature for 2 h to ensure complete dissolution of lignin. Insoluble impurities are removed by vacuum filtration to obtain a pure lignin solution.

The phenolic modification reaction is shown in Eq. 1.

$$\text{Lignin} + \text{Phenol} \xrightarrow{H_2SO_4} \textit{Phenolated Lignin} + H_2O \quad (1)$$

The pretreated lignin solution is mixed with phenol at a mass ratio of 1:1, and concentrated sulfuric acid is added as a catalyst. The amount of the catalyst is 5% of the mass of the lignin.

The mixture is stirred and reacted at 90 °C for 4 h. During the response, the phenol molecule replaces the hydrogen atoms on the aromatic ring in the lignin molecule through an electrophilic substitution reaction to generate phenolated lignin. After the reaction, the mixture is cooled to room temperature and diluted with deionized water. Its pH is adjusted to neutral to terminate the reaction. The phenolated lignin precipitate is obtained by centrifugal separation and repeatedly washed with deionized water until the liquid is neutral. The washed phenolated lignin is vacuum-dried at 60 °C for 24 h to obtain a dry phenolated lignin solid.

To further enhance the reactivity and polarity of lignin-based adhesives, this study further subjected phenolated lignin to a hydroxymethylation reaction with formaldehyde based on phenolic modification. The newly added phenolic hydroxyl group (–OH) in phenolated lignin molecules provides more reaction sites for subsequent hydroxymethylation.

The hydroxymethylation reaction refers to the process of introducing hydroxymethyl groups into lignin molecules to further improve their reactivity and bonding properties (Franco *et al*. 2021; Peralta *et al*. 2024). Industrial lignin is dissolved in deionized water at a concentration of 20 wt.%. Then, it is stirred at room temperature for 1 h to ensure that the lignin is completely dissolved. The hydroxymethylation reaction is shown in Eq. 2.

$$\text{Lignin} + CH_2O \xrightarrow{NaOH} \textit{Hydroxymethylated Lignin} + H_2O \quad (2)$$

Formaldehyde (37 wt.%) is added to the lignin solution. The molar ratio of formaldehyde to lignin is 1:1. Sodium hydroxide is added as a catalyst, and the amount of the catalyst is 5% of the lignin mass. Then they are stirred and reacted at 70 °C for 3 h. During the reaction, formaldehyde molecules undergo electrophilic addition reaction with the aromatic rings in the lignin molecules to generate hydroxymethylated lignin.

After the reaction is completed, the mixture is cooled to room temperature, and the pH is adjusted to neutral with dilute hydrochloric acid to terminate the reaction. Hydroxymethylated lignin precipitate is obtained by centrifugal separation and repeatedly washed with deionized water until the washing liquid is neutral. The washed hydroxymethylated lignin is vacuum-dried at 50 °C for 24 h to obtain a dry hydroxymethylated lignin solid.

To further enhance the performance of lignin-based adhesives, blending technology was employed to develop high-performance composite systems. In the first step, the

modified lignin was blended with polyvinyl alcohol (PVOH), followed by incorporation with epoxy resin. PVOH, a water-soluble polymer with good adhesion and film-forming properties (Gautam *et al.* 2022; Zhao *et al.* 2022), was dissolved in deionized water at a mass fraction of 10% and stirred at 90 °C for 2 h until fully dissolved. The prepared lignin was then dissolved in a 1,4-dioxane/water mixed solvent (volume ratio 7:3) at a concentration of 15 wt%, stirred at 60 °C for 1 h to ensure complete dissolution. Subsequently, the lignin solution was mixed with the PVOH solution in varying mass ratios (1:1, 1:2, 1:3, 2:1, and 3:1), followed by stirring at 80 °C for 4 h to ensure homogeneous blending of the components.

RESULTS AND DISCUSSION

Spectral Analysis

The structures of phenolated lignin and hydroxymethylated lignin were characterized by Fourier transform infrared spectroscopy (FTIR) (Magalhães *et al*. 2021; Liu and Kazarian 2022) and proton nuclear magnetic resonance (1H-NMR) (Gunawan *et al*. 2021; Riley *et al*. 2022) to confirm the successful progress of the reaction. The FTIR and 1H-NMR data are shown in Tables 1 and 2, respectively. In Table 1, the changes in the vibration peaks of hydroxyl groups, aromatic rings, and C-O bonds at different wave numbers reveal the effect of the hydroxymethylation process on the structure of lignin. Table 2 shows that the atomic signals of phenolated lignin and hydroxymethylated lignin at different chemical shifts were enhanced, which confirms the successful progress of the reaction.

Table 1. FTIR Data

Wavenumber (cm^{-1})	Absorption Peak Description	Phenolated Lignin	Hydroxymethylated Lignin
3400	Hydroxyl stretching vibration peak	Significantly enhanced	Further enhanced
3200-3600	Hydroxyl stretching vibration peak	-	Significantly enhanced
1600	C=C stretching vibration of aromatic rings	Weakly enhanced	Strongly enhanced
1050	C-O stretching vibration	Significantly enhanced	Significantly enhanced

Table 2. 1H-NMR Data

Chemical Shift (ppm)	Signal Description	Phenolated Lignin	Hydroxymethylated Lignin
6.5 - 7.5	Aromatic ring hydrogen signal	Enhanced	-
4.5 - 5.0	Hydroxymethyl hydrogen signal	-	Significantly enhanced

Mechanical Properties

Phenolated lignin and formaldehyde were mixed in a certain proportion to prepare phenolated lignin adhesive. Hydroxymethylated lignin and isocyanate were mixed in a certain proportion to prepare hydroxymethylated lignin adhesive. The tensile shear strength test (Zhou *et al*. 2021; Rajarajan *et al*. 2022) (ASTM D1002 2019) was used to compare the chemical modification method and evaluate the bonding performance of phenolated lignin adhesive and hydroxymethylated lignin adhesive on different wood substrates (oak and pine). Tensile shear strength test was calculated as follows,

$$\sigma = \frac{F}{A}(\mathrm{MPa}) \quad (3)$$

where F refers to shear force (N), and A refers to bonding area (mm²).

The two modification methods complement each other. Phenolic modification enhances the chemical reactivity of the adhesive. The hydroxymethylation reaction further improves its polarity and bonding properties. The combined application of these two modification methods can significantly improve the bonding strength of lignin-based adhesives.

Performance evaluation of adhesives at different blending ratios (modified lignin: PVOH): tensile shear strength test, water immersion test (ISO 7972 2023). The bonded samples were immersed in water for 24 h, and the retention rate of the bonding strength were measured, dynamic mechanical analysis (DMA) (Bashir 2021; Jani *et al*. 2021) (to evaluate the flexibility and viscoelasticity of adhesives). The results are shown in Fig. 1.

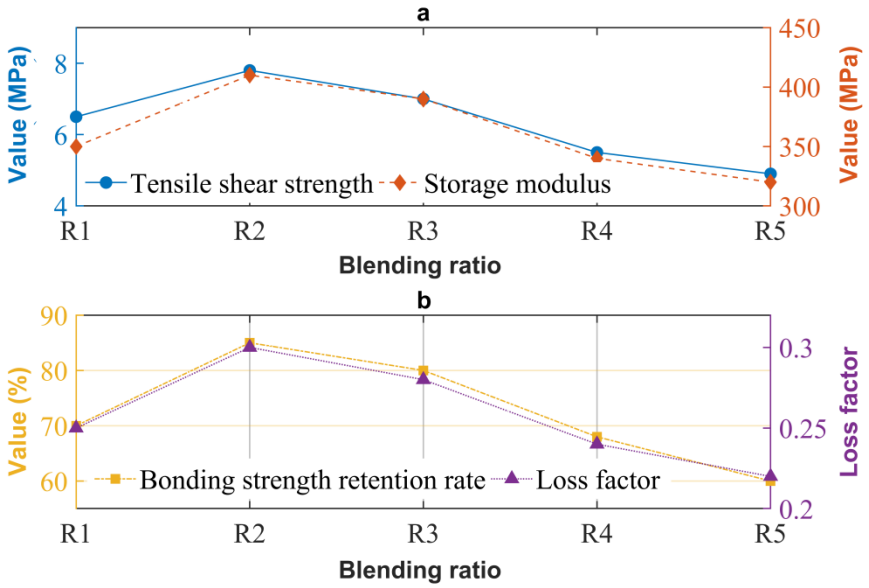

Fig. 1. Performance evaluation of adhesives at different blending ratios with PVOH. (a). Tensile shear strength and storage modulus; (b) bonding strength retention rate and loss factor. R1 to R5 refer to different blending ratios, specifically, 1:1, 1:2, 1:3, 2:1, and 3:1.

When the mass ratio of modified lignin to PVOH was 1:2, the bonding strength retention rate of the adhesive reached 85%. The tensile shear strength reached 7.8MPa, and the storage modulus and loss factor were higher than those of other blending ratios, indicating that its flexibility and viscoelasticity are improved.

Epoxy resin is a high-performance thermosetting resin with excellent bonding strength, chemical corrosion resistance, and thermal resistance. Bisphenol A epoxy resin (E-51) was dissolved in acetone at a mass fraction of 20% and stirred at room temperature for 1 h until the epoxy resin is completely dissolved. The modified lignin solution was mixed with the epoxy resin solution in different mass ratios (same as PVOH) and stirred at 60 °C for 3 h to ensure that the two components were fully mixed.

Triethylenetetramine was added as a curing agent, and the amount of the curing agent is 10% of the mass of the epoxy resin. After the mixture is stirred evenly, it is applied to the wood surface and cured at 120 °C for 2 h to obtain a blended adhesive bonding sample.

The bonding performance of adhesives at different blending ratios (modified lignin: epoxy resin) was evaluated through tensile shear strength test, thermal gravimetric analyzer (TGA, to evaluate the thermal resistance of the adhesive), and DMA. The results are shown in Fig. 2. When the mass ratio of modified lignin to epoxy resin was 1:2, the tensile shear strength of the adhesive reached 9.5 MPa. The blended adhesive's storage modulus and loss factor were higher than those of PVOH with other blending ratios. Moreover, the initial decomposition temperature of the adhesive at this ratio reached 280 °C, and the thermal resistance was significantly improved. In summary, modified lignin was compounded with PVOH and epoxy resin through blending technology to prepare an adhesive with excellent performance.

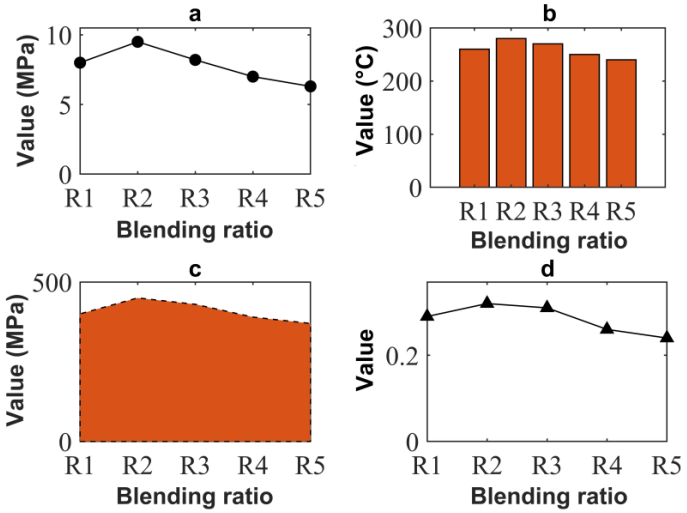

Fig. 2. Performance evaluation of adhesives at different blending ratios with epoxy resin. (a) Tensile shear strength; (b) initial decomposition temperature; (c) storage modulus; (d) loss factor

Low temperature and low-pressure production process

The combination of microwave-assisted heating and mechanical stirring technology is a low-temperature and low-pressure production process that synthesizes adhesives under low temperature (<100 ° C) and low-pressure conditions, which can reduce the production energy consumption and cost of lignin-based adhesives (Wang *et al.* 2022).

Microwave-assisted heating is an efficient and fast heating method that can achieve uniform heating in a short time, reduce heat loss, and improve energy efficiency (Devi *et al*. 2021; Gao *et al*. 2021). Equation 4 shows formula for the microwave heating energy efficiency model,

$$\eta_{microwave} = \frac{P_{absorbed}}{P_{input}} \times 100\% = \frac{\varepsilon'' \cdot f \cdot E^2 \cdot V}{P_{input}} \times 100\% \quad (4)$$

where ε'' refers to the dielectric loss factor (0.35 for the 1,4-dioxane/water system), f refers to the microwave frequency (2.45GHz), E^2 refers to the electric field intensity (V/m), and V refers to the volume of the reaction system (2 L).

Although microwave-assisted heating technology has shown significant advantages in reducing energy consumption and shortening reaction time, its industrial promotion may face practical challenges. Traditional adhesive production lines often use mature processes such as oil bath heating or steam heating, which result in high equipment renovation costs. Additionally, microwave reactors require specific frequency (2.45 GHz) and power control technology, which may increase initial investment. In scenarios where microwave equipment cannot be used, traditional processes are still feasible, but a balance needs to be struck between efficiency and cost.

The non-isothermal reaction kinetics equation is calculated according to the Arrhenius modification model, as follows,

$$\frac{d\alpha}{dt} = A \cdot e^{-\frac{E_a}{R(T+\Delta T_{mic})} \cdot (1-\alpha)^n} \quad (5)$$

where α refers to the reaction conversion, ΔT_{mic} refers to the microwave thermal effect of additional temperature (about 15 °C), E_a refers to the apparent activation energy (45kJ/mol for hydroxymethylation reaction), and n refers to the reaction order (level 1.5).

A microwave-transparent closed reactor with a volume of 2 liters and made of corrosion-resistant polytetrafluoroethylene (PTFE) was selected. The reactor has a built-in stirring device and a temperature sensor for real-time monitoring and control of the reaction temperature.

A single-mode microwave reactor was used with a maximum power of 1,000W. The microwave generator was connected to the reactor through a waveguide to ensure efficient transmission of microwave energy. Insulation materials were wrapped around the outside of the reactor to reduce heat loss and improve heating efficiency.

The modified lignin, solvent (1,4-dioxane/water mixed solvent), and catalyst (sulfuric acid) were added to the reactor in proportion. The microwave heating system was started. The heating power was set to 600 W. The reaction temperature was monitored and controlled in real time by a PID (Proportional Integral Derivative) (Shi *et al*. 2022) controller to ensure that the temperature was within the set range. The surface temperature of the reactor was monitored in real time by an infrared temperature sensor to prevent local overheating and ensure uniform heating.

Mechanical stirring is the key to ensuring that the reactants are fully mixed and react evenly. The stirring power coefficient correlation adopted the Rushton turbine correction model, as follows,

$$N_p = \frac{P}{\rho N^3 D^5} = 0.85\left(\frac{D}{T}\right)^{-0.3}\left(\frac{H}{T}\right)^{0.6} \quad (6)$$

where N_p refers to the power number (0.35 after optimization), D refers to the blade diameter (140 mm), T refers to the reactor diameter (200 mm), and H refers to the liquid level (180 mm).

The complete mixing of reactants in the reactor required the use of mechanical stirring technology, with anchor type stirring blades installed inside the reactor. The diameter of the stirring blade was 70% of the inner diameter of the reactor.

The stirring paddle was made of stainless steel and coated with PTFE to prevent

corrosion and adhesion. The stirring speed was controlled by a variable frequency speed motor with a stirring speed range of 100 to 800 rpm.

The stirring speed was adjusted according to the viscosity of the reaction system and the reaction stage. In the early stage of the reaction, the stirring speed was set to 600 rpm to ensure rapid mixing of the reactants. In the later stage of the reaction, the stirring speed was reduced to 300 rpm to reduce energy consumption and prevent side reactions caused by excessive stirring. Through computational fluid dynamics (CFD) simulation, the stirring paddle design and stirring speed were optimized to ensure that the reactants are evenly mixed in the reactor.

Compared with traditional mechanical stirring, the optimization parameters and energy consumption analysis are shown in Table 3. The diameter ratio of the traditional stirring paddle was 50%, while that for the optimized stirring paddle was increased to 70%, which indicates that the mixing efficiency can be improved by increasing the diameter of the paddle blade. In terms of rotation speed, the optimized stirring allowed adjustment within the range of 100 to 800 rpm to meet different mixing requirements, and the mixing uniformity was increased from 82% to 95%. CFD simulation requires that the Reynolds number be greater than 10^4 to ensure that the flow state is turbulent and to obtain more accurate simulation results. At the same time, the unit energy consumption of optimized stirring was reduced to 0.56 kWh/h, showing a significant improvement in energy efficiency. These data show that optimized stirring not only improved mixing uniformity and energy efficiency but it also met the strict conditions of CFD simulation.

Adhesive synthesis was carried out under low-temperature and low-pressure conditions. A PID temperature controller is used, combined with microwave heating and mechanical stirring, to achieve precise control of the reaction temperature. The analysis of process parameter control precision is shown in Table 4.

According to the data in Table 4, the set temperature control value was 80 °C. The control precision reached ±1 °C. The fluctuation range was only ±0.8 °C. The parameters of the PID adjustment algorithm used reflect the system's sensitivity and reaction speed to temperature changes. In terms of pressure control, fuzzy PID control was used to adapt to pressure changes. The stirring torque was set to 0.65 ± 0.05 N·m, with a fluctuation range of ±0.03 N·m. Feedforward-feedback composite control was applied to ensure a stable stirring effect. The set microwave power value was 600 ± 20W, with a fluctuation range of ±15W. Using adaptive predictive control improved the flexibility of power regulation.

Table 3. Mechanical Stirring Optimization Parameters and Energy Consumption Analysis

Parameter	Traditional Stirring	Optimized Stirring	CFD Simulation Constraints
Paddle diameter ratio	50%	70%	Reynolds number Re > 10^4
Speed range (rpm)	Fixed at 600	Adjustable 100-800	Turbulence intensity > 0.8
Mixing uniformity (%)	82	95	Variance $\sigma^2 < 0.05$
Unit energy consumption (kWh/h)	0.80	0.56	Power factor cosφ = 0.92
Shear rate (s^{-1})	120	Adjustable 80-200	Weissenberg number Wi < 5

Table 4. Process Parameter Control Precision

Control Parameter	Set Value	Fluctuation Range	PID Control Algorithm
Temperature control (°C)	80±1	±0.8	Kp=2.5, Ki=0.3, Kd=1.2
Pressure control (kPa)	Atmospheric ±5	±3.2	Fuzzy PID control
Stirring torque (N·m)	0.65±0.05	±0.03	Feedforward-feedback composite control
Microwave power (W)	600±20	±15	Adaptive predictive control

During the reaction, the reaction temperature was monitored in real time by a thermocouple sensor, and the microwave heating power and stirring speed are dynamically adjusted according to the temperature changes to ensure that the temperature was stable within the set range (<100 °C).

A pressure sensor was installed on the reactor to monitor the reaction pressure in real time. The pressure of the reaction system was controlled at normal pressure or slightly negative pressure by a back pressure valve and a vacuum pump to avoid pressure increase caused by temperature increase. In the hydroxymethylation reaction, the reaction temperature was controlled below 80 °C by controlling the pressure in the reactor, effectively avoiding the occurrence of side reactions.

The reaction endpoint was determined by online infrared spectroscopy (IR) monitoring, which monitors the changes of key functional groups in the reaction system in real time to determine the reaction endpoint. In the phenolation, the completion of the reaction is determined by monitoring the changes in the absorption peaks of -OH and -COOH.

Table 5. LCA System Boundary and Functional Unit Definitions

Element	Parameter Setting	Remarks
System boundary	Agricultural and forestry stage → Chemical processing → Production → Use → Waste disposal	Closed system, covering the entire life cycle
Functional unit	1m³ plywood bonding operation	Comparison benchmark: Petroleum-based adhesives
Distribution method	Mass distribution method (Lignin:Other components = 1:9)	Handling the environmental load of co-products
Time frame	20 years (including raw material planting period)	Considering the biological carbon dynamic sequestration effect

Application of life cycle assessment method

Based on ISO 14040/44 (2006), a life cycle assessment model was established for lignin-based adhesives to quantify their environmental benefits and identify key influencing factors (Sala *et al.* 2021). The LCA system boundary and functional unit

definitions are shown in Table 5. The system boundaries in Table 5 cover the stages to form a closed system from cradle to grave. The bonding operation is performed on 1 m³ plywood to ensure lateral comparability with petroleum-based adhesives. The mass distribution method is used to treat the lignin co-product (pulping black liquor), and the environmental load is distributed according to the mass ratio (lignin: other components=1:9).

Taking wheat straw lignin as a benchmark case, agricultural input data of typical farmland in North China were collected (fertilizers: urea 220 kg/ha; phosphate fertilizer 150 kg/ha; agricultural machinery diesel oil consumption 85 L/ha; irrigation water 450 m³/ha), and the global average transportation distance (road transportation 200km) was linked through the Ecoinvent 3.8 database.

Blending components: The PVOH used is the average production data of the petrochemical industry (ethylene process, with an energy consumption of 38 MJ/kg), and the epoxy resin used was the upstream chlorohydrin process data (with a bisphenol A consumption of 0.85 kg/kg).

The reactor energy consumption was calculated by combining Aspen Plus simulation (with a microwave heating power density of 15 W/g and a thermal efficiency of 82%). The mechanical stirring process was simulated by the discrete element method, and the unit output energy consumption (0.56 kWh/kg) was calculated, with a deviation of <5% from the measured data.

VOC emissions were measured using gas chromatography-mass spectrometry (GC-MS) to detect volatile organic compounds (formaldehyde, benzene series) released during the curing process of the adhesive and a release kinetic model was established in combination with Fick's diffusion law, as follows,

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - kC \quad (7)$$

where the diffusion coefficient is $D = 2.3 \times 10^{-8} m^2/s$, and the degradation rate constant is $k = 0.015 h^{-1}$.

Three scenarios were set for waste treatment: incineration (with a calorific value of 18 MJ/kg and a flue gas purification efficiency of 95%), landfill (with a methane yield of 0.12 m³/kg and a collection rate of 60%), and mechanical recycling (with an energy consumption of 1.2 kWh/kg).

The environmental impact assessment adopts the method of combining point source and midpoint in ReCiPe 2016 (Hierarchist version). The LCA model was built based on SimaPro 9.3. The Brigade model was integrated to process agricultural stage data, and the GaBi extension package was used to process chemical process data. Ten thousand iterations were set to analyze the sensitivity of key parameters to the results. The time range was set to 20 years, and the key parameters of the sensitivity analysis are shown in Fig. 3.

The fluctuation range of microwave heating energy consumption was ±15%. Its impact weight on carbon footprint was 23%, and its impact weight on VOC emissions was only 5%. This shows that microwave heating energy consumption was more important in carbon emissions than its impact on VOC release. Secondly, the fluctuation range of the VOC release rate was ±20%. Its impact weight on carbon footprint was 8%, and its impact weight on VOC emission was as high as 68%, showing its criticality in controlling VOC emission. The waste recovery rate ranged from 20% to 50%, and the corresponding impact weights on carbon footprint and VOC emission were 18% and 12%, respectively, indicating that increasing the waste recovery rate helped to reduce carbon emissions but

had a relatively small impact on VOC emissions. Finally, the fluctuation range of catalyst dosage was ±10%. Its impact weight on carbon footprint was 12%, and its impact weight on VOC emissions was 22%, which means that the use of catalysts plays a greater role in reducing VOC emissions.

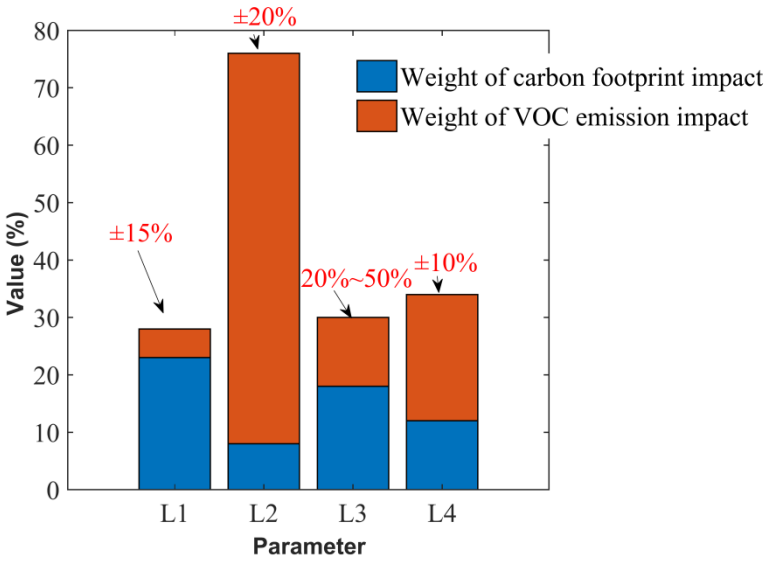

Fig. 3. Key parameters of sensitivity analysis. L1 to L4 refer to microwave heating energy consumption, VOC release rate, waste recovery rate, and catalyst dosage, respectively.

CRITICAL ANALYSIS

The comparison results of the chemical modification methods are shown in Fig. 4. The tensile shear strength of the phenolated lignin adhesive reached 6.5 MPa, which is about 40% higher than that of the unmodified lignin adhesive. This is mainly attributed to the introduction of a large number of active groups by the phenolation, which enhances the interaction between the adhesive and the wood substrate.

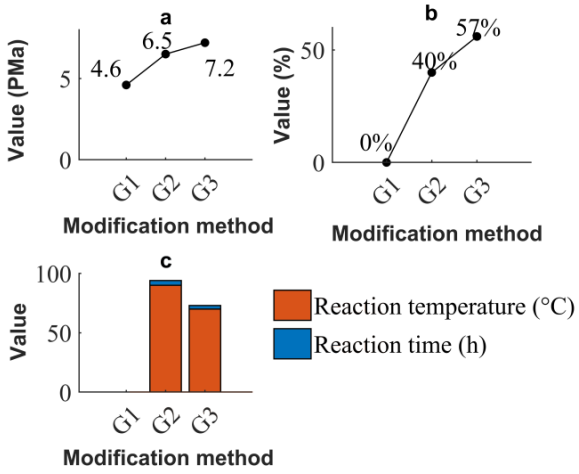

Fig. 4. Comparison of chemical modification methods. (a) Effect of modification method on tensile shear strength; (b) bonding strength improvement rate; (c) reaction temperature and reaction time. G1 to G3 represent without modification, with phenolic modification, and with hydroxymethylation reactions, respectively.

The tensile shear strength of the hydroxymethylated lignin adhesive reached 7.2 MPa, which is about 57% higher than that of the unmodified lignin adhesive. This is mainly attributed to the introduction of a large number of polar groups by the hydroxymethylation reaction, enhancing the interaction between the adhesive and the wood substrate.

A comparison of some of the process parameters of microwave-assisted heating and traditional oil bath heating is shown in Fig. 5. Compared with traditional oil bath heating, microwave-assisted heating shortened the heating time by 63% and reduces energy consumption by 40% at the same temperature. This is of great significance for energy conservation and emission reduction. Under the same temperature, microwave heating only took 15 min for hydroxymethylation reaction, while oil bath heating took 40 min. In terms of energy efficiency, microwave heating reached 85%, which was 63% higher than traditional oil bath heating. Finally, the risk of local overheating was less than 5% in microwave heating, while it was as high as 35% in traditional methods. The former reduced the risk by 86%.

The high efficiency of microwave heating can be mainly attributed to its selective heating characteristics. Microwave energy directly acts on polar molecules (such as water, solvents, and reactants), reducing the heating of non-polar molecules, and thereby improving energy utilization efficiency.

The lignin-based adhesive synthesized by the low-temperature and low-pressure production process constructed in this paper is compared with the traditional process products, as shown in Table 6. In terms of tensile shear strength, the value of the process in this paper is 8.5 ± 0.3 MPa, which is significantly higher than the 7.7 ± 0.5 MPa of the traditional process, indicating that the improved process can provide stronger bonding. In addition, the bonding retention rate is also improved. The process of this paper reached 90 ± 2%, while the traditional process was only 75 ± 4%. Thus, the present result met the ISO 9022-12 (2015) standard and showed better durability and reliability. The process of this paper was judged to be superior to the traditional process in multiple performance indicators, showing its potential and value in practical applications.

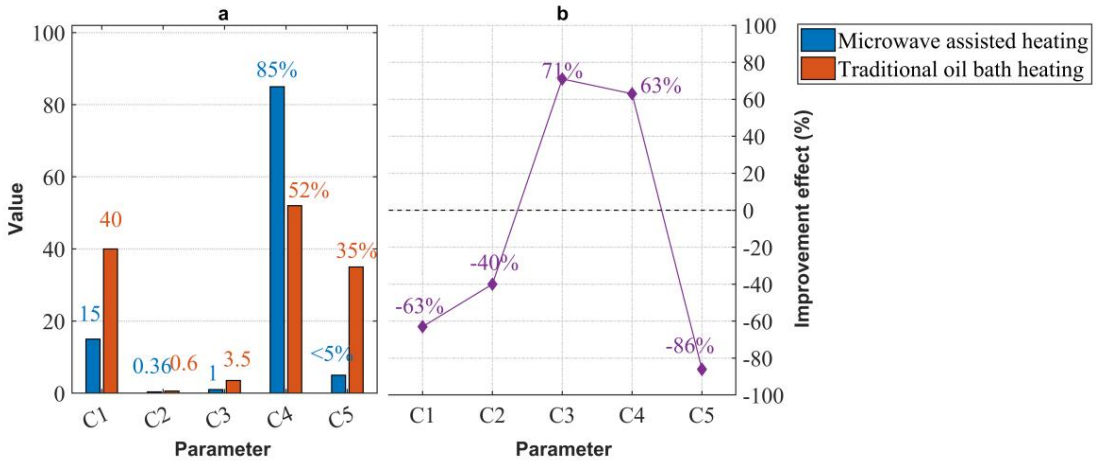

Fig. 5. Comparison of process parameters between microwave-assisted heating and traditional oil bath heating. (a). Specific data of process parameter comparison; (b) improvement effect of process parameter comparison. C1 to C5 refer to heating time (min), energy consumption (kWh/kg), temperature uniformity (°C), energy efficiency (%), and local overheating probability, respectively. Negative values indicate reduction or shortening, and positive values indicate improvement or enhancement.

Table 6. Product Performance Comparison and Verification

Performance Indicator	This Process	Traditional Process	Testing Standard
Tensile shear strength (MPa)	8.5 ± 0.3	7.7 ± 0.5	ASTM D1002 (2019)
Bonding retention rate (%)	90 ± 2	75 ± 4	ISO 9022-12 (2015)
Storage modulus (MPa)	480 ± 20	350 ± 30	DMA 1Hz
Loss factor	0.28 ± 0.02	0.35 ± 0.03	ISO 6721-1 (2019)
Storage stability (Months)	> 6	3	Accelerated test at 40 °C

Through LCA analysis, the comparison between the lignin-based adhesive prepared in this paper and the traditional petroleum-based adhesive is shown in Fig. 6. The carbon footprint of the lignin-based adhesive was 0.8 kg CO_2-eq/kg, which is much lower than the 1.33 kg CO_2-eq/kg of the traditional petroleum-based adhesive, with an improvement of 40%. This shows that the use of the lignin-based adhesive can effectively reduce greenhouse gas emissions and reduce the impact on the environment. In addition, in terms of VOC emission, the emission of the lignin-based adhesive was only 0.18 kg, while that of the traditional adhesive was 0.36 kg, with an improvement of 50%, showing its positive impact on air quality. In terms of resource consumption, the resource consumption of the lignin-based adhesive was 6.0 kg, compared with 8.57 kg of the traditional adhesive, with an improvement of 30%, indicating that the lignin-based adhesive can use resources more efficiently and reduce the demand for raw materials. Finally, in terms of waste generation, the lignin-based adhesive generated 0.46 kg of waste, while the traditional adhesive generated 0.75 kg, with an improvement of 38.67%. Overall, these data show that the lignin-based adhesive has obvious advantages in reducing environmental burden and improving resource utilization efficiency, which meets the current requirements of sustainable development. It is gaining more and more attention and application.

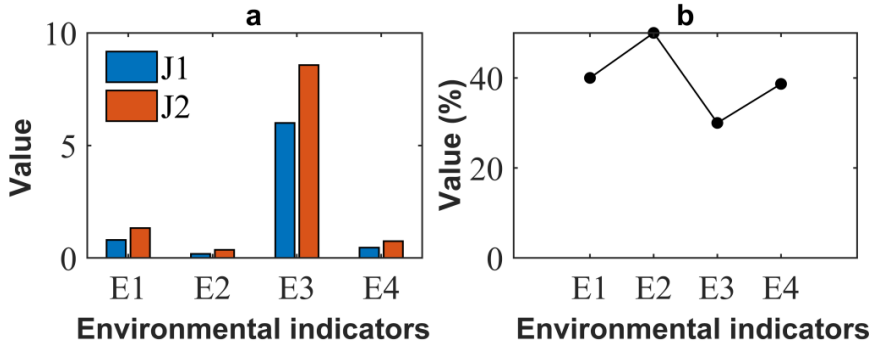

Fig. 6. Comparison between lignin-based adhesives and traditional petroleum-based adhesives. (a) Comparison of environmental indicators; (b) improvement rate of environmental indicators. E1 to E4 represent carbon footprint (kg CO_2-eq/kg), VOC emission (kg), resource consumption (kg), and waste generation (kg), respectively.

Although significant achievements have been made in the research, development, and application of lignin-based adhesives, there are still some shortcomings. The complexity of the production process and cost control still need further optimization, especially in large-scale industrial production, so balancing cost and performance remains

a challenge. In the future, research should focus on developing more efficient and economical production processes. In addition, there is a need to further validate the stability and durability of lignin-based adhesives under different environmental conditions by expanding sample sizes and extending experimental periods.

CONCLUSIONS

1. The application of chemical modification and blending technology has enabled the successful preparation of high-performance and low-cost lignin-based adhesives and verified their feasibility and superiority in wood processing. The low-temperature and low-pressure production process effectively reduces production costs and energy consumption.
2. Life cycle assessment (LCA) results showed that lignin-based adhesives have significant environmental benefits in terms of carbon footprint and volatile organic carbon (VOC) emissions.
3. The modified adhesives were found to be comparable to traditional petroleum-based adhesives in terms of adhesive strength, heat resistance, and environmental adaptability, meeting the industrial standards for engineering wood (plywood, particleboard).
4. Large-scale application of the technology is still limited by differences in raw material batches and equipment modification costs. Future research needs to focus on standardizing raw material pretreatment, dynamically regulating cross-linking networks, and coupling microwave technology with other green technologies to balance performance, cost, and ecological benefits.
5. This paper demonstrates that through multi-scale collaborative optimization, lignin-based adhesives not only can replace traditional resins, but they also can promote the transformation of wood processing towards closed-loop sustainable manufacturing.

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Author Contributions

Xueping Qiu: software and testing. Yanran Sun: data curation and investigation. Xingxian Lan: analyzing the data. The manuscript was written through the contributions of all authors. All authors have given their approval to the final version of the manuscript.

Data Availability

The datasets used or analyzed during the current study are available from the corresponding author upon reasonable request.

Competing Interests

The authors declare that they have no competing interests regarding the publication of this paper.

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