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Lignin-Based Adhesives: Linking Isolation History and Molecular Structure to Interfacial and Bulk Performance

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Abstract: Conventional synthetic adhesives are indispensable to modern manufacturing, yet many depend on nonrenewable feedstocks and some contain constituents of health or environmental concern. Lignin is an abundant renewable aromatic polymer with phenolic and aliphatic hydroxyl groups, an aromatic framework, and potential antioxidant activity. These features support covalent curing, intermolecular association, and interfacial interactions in adhesive formulations. This review examines how biomass origin, isolation history, fractionation, and chemical modification influence lignin structure, reactivity, solubility, compatibility, curing behavior, and interfacial and bulk performance. It separates adhesion at the substrate interface from cohesion within the cured network and evaluates trade-offs among bonding performance, processability, feedstock variability, reagent and solvent demand, and environmental benefit. The analysis identifies the material specifications, test controls, and processing requirements needed for reproducible, scalable, and commercially credible lignin-based adhesives.

Keywords: technical lignin, isolation history, structure–property relationships, curing chemistry, sustainable adhesives

Introduction

High-performance adhesives are used in wood products, packaging, transportation, electronics, and construction. Epoxy, melamine-formaldehyde, phenol-formaldehyde, and polyurethane are among the principal chemistries in these applications.[1-3] Many rely on nonrenewable feedstocks, and some contain compounds of health or environmental concern. Bisphenol A, a common epoxy-resin precursor, is an endocrine-active chemical under regulatory scrutiny.[4,5] Formaldehyde released during manufacture or from bonded wood products raises occupational and indoor-air-quality concerns and is subject to strict emission limits.[6,7] These constraints have increased interest in adhesives derived wholly or partly from renewable resources.

Most previous reviews classify lignin-based adhesives by resin chemistry, modification route, or application.[8-11] This review instead treats isolation history as a primary variable in adhesive performance.[10,11] Isolation controls molar-mass distribution, functional-group content, condensation, residual impurities, and solubility,[12,13] whereas formulation, curing, substrate, and test protocol also determine measured bond strength.[8,10,11] Cross-study comparison is meaningful only when these variables are considered together and interfacial adhesion is distinguished from cohesion within the cured adhesive.

Industrial pulping and biorefinery operations generate large quantities of technical lignin. Native lignin is an irregular phenylpropanoid polymer joined mainly by aryl-ether and carbon-carbon linkages. Technical lignins retain the aromatic framework and phenolic and aliphatic hydroxyl groups, but their structures and functional-group distributions vary with botanical origin and isolation.[14,15] Lignin can therefore act as a reactive resin component rather than an inert filler when its reactivity, solubility, heterogeneity, and matrix compatibility match the chosen formulation and substrate.

The review follows lignin from fractionation and purification through chemical modification, network formation, and application. It relates these operations to reactivity, compatibility, curing behavior, interfacial adhesion, and bulk cohesion. Wood bonding and emerging high-performance systems are assessed together with constraints imposed by feedstock variability, processability, reproducibility, and scale-up. Figure 1 summarizes the relationships among lignin

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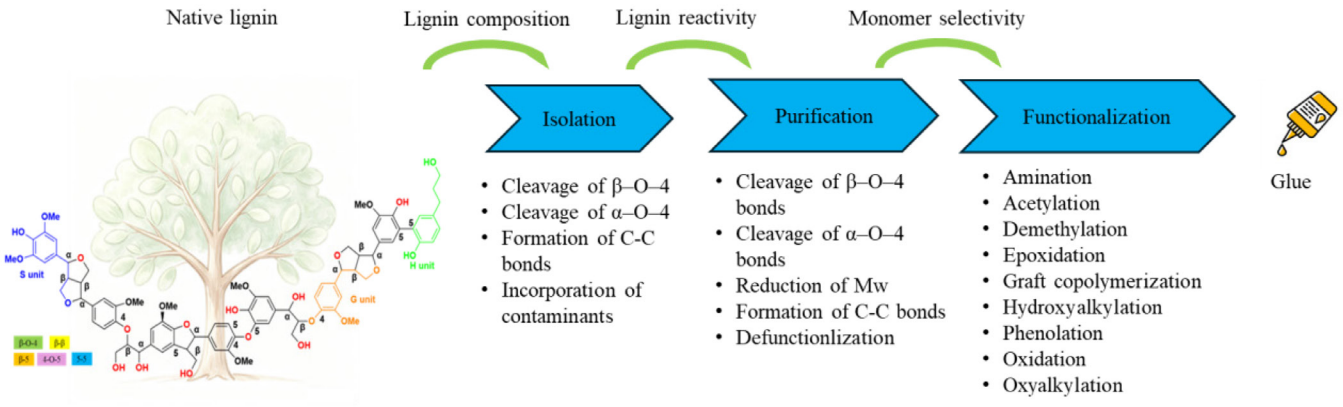

Figure 1. Scope of the review: lignin source and isolation, purification and structural characterization, chemical activation, network formation, and application-specific adhesive performance.

origin, isolation history, molecular structure, processing, and adhesive performance.

Structure and Composition of Lignin

1. Native Lignin Architecture and Biological Variability

In lignocellulosic biomass, lignin is deposited predominantly in secondary cell walls and is present at particularly high concentrations in the compound middle lamella and cell corners.[16] There, it contributes to cell-wall rigidity, hydrophobicity, and resistance to biological degradation. Native lignin forms an irregular phenolic network associated with cellulose and hemicelluloses. It contains phenolic hydroxyl, aliphatic hydroxyl, and methoxy groups, together with smaller amounts of carbonyl and carboxyl groups. Lignin is biosynthesized primarily through oxidative radical coupling of three monolignols, *p*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol, which give rise to *p*-hydroxyphenyl (H), guaiacyl (G), and syringyl (S) units, respectively.[17] The arylglycerol-*β*-aryl ether (*β*-*O*-4) is generally the most abundant interunit linkage, accompanied by resinol (*β*-*β*), phenylcoumaran (*β*-*5*), biphenyl (5-5′), dibenzodioxocin (5-5′/*β*-*O*-4), and spirodienone (*β*-1/*α*-*O*-*α*) structures. Other lignin architectures may contain additional structural motifs, including benzodioxane units characteristic of C-lignin and tricin-derived units found predominantly in grass lignins.[18]

Lignin composition and interunit-linkage distribution vary with botanical origin, tissue type, developmental stage, and growth conditions.[19-21] Softwood lignins are generally rich in G units, whose unsubstituted C5 positions favor the formation of condensed carbon–carbon linkages, particularly *β*–5 and 5–5′ linkages. Hardwood lignins contain both G and S units and commonly exhibit higher *β*-*O*-4 contents and lower degrees of condensation as the S/G ratio increases. Grass lignins contain variable proportions of H, G, and S units together with hydroxycinnamate moieties, particularly *p*-coumarate and ferulate moieties, that occur in ester- or ether-linked forms. These compositional tendencies affect the response of lignin to cleavage, fractionation, and functionalization. However, botanical origin alone does not determine thermal stability, antioxidant activity, water resistance, or adhesive performance because these properties are also strongly influenced by isolation history, molar-mass distribution, phenolic hydroxyl content, residual impurities, and formulation conditions.

Isolation converts the native network into heterogeneous technical lignin fragments with altered molar-mass distribution, functionality, ether-linkage content, condensation, and impurity profile. Conditions should therefore be selected for the intended adhesive chemistry, retaining or generating useful functionality while limiting uncontrolled condensation, degradation, and contamination.

2. Isolation and Pretreatment as Structural Design Variables

Lignin isolation relies primarily on chemical or solvent-based fractionation, whereas physical, physicochemical, and biological treatments generally assist biomass deconstruction or modify lignin before recovery. These processes affect lignin yield, purity, molar-mass distribution, linkage composition, degree of condensation, functional-group content, and residual impurity levels.[22] The suitability of the recovered lignin must therefore be evaluated in relation to the intended

adhesive formulation rather than solely on the basis of lignin yield.

2.1 Chemical Fractionation

Chemical and solvent-based fractionation methods use alkaline, acidic, sulfite-based, organosolv, ionic liquid, or deep eutectic solvent systems to disrupt lignin–carbohydrate associations and solubilize lignin.[22,23] The extent of structural alteration depends on reagent composition, temperature, reaction time, pressure, water content, and the method used to recover lignin from the resulting liquor.[24]

The kraft process remains the dominant chemical pulping route. Alkaline sulfide liquor cleaves aryl-ether linkages and solubilizes lignin as phenolate- and carboxylate-rich fragments in black liquor.[25] Recovered kraft lignin commonly shows extensive ether cleavage, condensation, broad molar-mass distribution, and sulfur-containing functionality.[26] These features can limit solubility and aromatic-site accessibility, although ether cleavage increases phenolic hydroxyl content and can improve reactivity in selected curing reactions. Its behavior must be interpreted with botanical source, pulping severity, and recovery process.

Soda delignification uses sodium hydroxide without sulfide and is particularly suitable for many non-woody feedstocks, including straw and bagasse, because of their relatively accessible structure and lower recalcitrance.[27] Soda lignin is sulfur-free, although its molar mass, degree of condensation, and impurity content depend strongly on pulping and precipitation conditions. Sulfite pulping employs sulfite or bisulfite salts of sodium, calcium, magnesium, or ammonium.[28] Sulfonation occurs together with ether-bond cleavage and condensation, producing lignosulfonates containing hydrophilic sulfonate groups, primarily at benzylic positions.[29] Their water solubility is advantageous for aqueous adhesive formulations, but their high hydrophilicity may impair water resistance unless they are effectively incorporated into the cured network.

Nitric acid treatment can remove or transform lignin at relatively low temperatures.[30] However, nitric acid may induce oxidation, nitration, and depolymerization and should therefore be regarded as an oxidative conversion process rather than a structure-preserving isolation route. More generally, acidic fractionation can cleave *α-O*-4 and *β-O*-4 ether linkages but may also generate benzylic carbocation intermediates that promote carbon-carbon condensation, particularly under severe conditions.[31,32] Acid treatment cannot therefore be assumed to minimize lignin condensation.

Organosolv fractionation uses an organic solvent, commonly with water and an acid or base catalyst. Under moderate conditions it can yield relatively pure lignin with greater *β-O*-4 retention than kraft lignin, whereas severe or strongly acidic conditions promote ether cleavage and condensation. Elevated temperature and pressure, solvent flammability, and solvent recovery remain important constraints.[25,33] Bio-derived solvents such as Cyrene™, *γ*-valerolactone, and dimethyl isosorbide have also been studied.[34,35] Their environmental value depends on solvent production, recycling, energy demand, and lignin recovery efficiency.

Ionic liquids and deep eutectic solvents provide compositionally tunable media for lignin fractionation. Imidazolium-based ionic liquids can disrupt lignin-carbohydrate interactions through anion-dependent hydrogen-bonding and solvation effects and may promote C-O bond cleavage under selected conditions.[36,37] Deep eutectic solvents are commonly formed from hydrogen-bond acceptors and donors, and their fractionation behavior depends strongly on composition, acidity, water content, and treatment severity.[38,39] Major barriers to scale-up include solvent cost, high viscosity, water management, product washing, solvent loss, and the energy required for solvent recovery.[40-42]

2.2 Physical and Physicochemical Treatments

Mechanical treatments such as ball milling[43] and ultrasonication[44] disrupt lignocellulosic structure, reduce particle size, and increase substrate accessibility. Depending on treatment intensity, they may also alter lignin molar mass or expose functional groups. However, these methods generally do not provide selective lignin isolation when used alone and may involve substantial energy consumption.

Microwave irradiation[45,46] is more accurately regarded as a rapid heating method used to intensify chemical or solvent-based fractionation than as an independent lignin-isolation mechanism. Homogenization[47,48] can increase surface area and improve mass transfer but likewise requires a subsequent separation or recovery step.

Physicochemical treatments combine thermal, mechanical, pressure, and solvent effects. Steam explosion,[49,50] hydrothermal treatment,[51] and supercritical fluid processing[52,53] have been investigated for biomass deconstruction and lignin recovery. Their effects depend strongly on treatment severity and may include lignin redistribution, improved accessibility, ether-bond cleavage, depolymerization, and condensation.

Microwave- or ultrasound-assisted deep eutectic solvent treatments[54-57] and extrusion-assisted deep eutectic solvent pretreatment[58,59] have been investigated to reduce processing time and improve mass transfer or fractionation efficiency. Their industrial relevance depends on total energy consumption, solvent recovery, equipment scalability, product purity, and whether the recovered lignin possesses properties suitable for the intended adhesive.

2.3 Biological Delignification and Enzymatic Conversion

Biological pretreatment employs ligninolytic fungi, bacteria, or oxidative enzymes to modify or remove lignin under comparatively mild conditions.[60-63] Relevant enzymes include laccases, lignin peroxidases, manganese peroxidases, versatile peroxidases, dye-decolorizing peroxidases, and unspecific peroxygenases. Their activity and selectivity depend on enzyme source, mediator system, substrate accessibility, pH, temperature, solvent environment, and mass transfer.

Biological delignification, enzymatic depolymerization, and recovery of polymeric lignin represent distinct objectives and should not be treated as equivalent isolation methods. Selective enzymatic transformation can introduce or expose useful functionality, whereas extensive depolymerization may reduce molar mass and the ability of lignin to contribute to the cohesive strength of an adhesive network unless the resulting fragments are subsequently functionalized or crosslinked. Long residence times, enzyme cost and stability, mass-transfer limitations, and product heterogeneity remain major barriers to direct implementation in adhesive manufacture.

Table 1 summarizes the structural tendencies of representative fractionation and pretreatment routes. A waterborne adhesive may benefit from hydrophilic functionality, whereas a structural thermoset may require low impurity content, controlled molar-mass distribution, limited hydrophilicity, and accessible reactive sites.

Molecular Origins of Interfacial Adhesion and Bulk Cohesion

Adhesive-joint strength and durability reflect interfacial adhesion, cohesion within the cured adhesive, and sometimes substrate integrity. Interfacial mechanisms include wetting,

Table 1. Qualitative Effects of Representative Lignin Fractionation and Pretreatment Routes on Properties Relevant to Adhesive Formulation

Method	Representative feedstock	Potential advantage	Principal limitation	Typical structural implication	Ref.
Alkaline pretreatment	Lignocellulosic biomass	Effective lignin removal; sulfur-free lignin can be obtained in soda-based processes	Chemical recovery, wastewater generation, and variable impurity content	Ether-bond cleavage and changes in phenolic hydroxyl content; condensation depends on treatment severity and recovery conditions	64
Organosolv fractionation	Bamboo and agricultural residues	Potentially high lignin purity, low sulfur content, and tunable structural preservation	Solvent flammability, pressure requirements, cost, and solvent recovery	β-O-4 retention and hydroxyl accessibility depend strongly on solvent composition and treatment severity	64,65
Ionic-liquid fractionation	Lignocellulosic biomass	High lignin solubility and tunable solvent chemistry	Cost, viscosity, washing demand, solvent recovery, and solvent-specific toxicity concerns	Lignin dissolution and C–O bond cleavage depend on ion pair, water content, and process severity	66,67
Deep-eutectic-solvent fractionation	Coniferous lignocellulosic biomass	Tunable solvent composition and the possibility of coupling delignification with component separation	High viscosity, water management, product washing, and solvent recovery	Molar mass, condensation, and hydroxyl-group accessibility vary with DES composition and treatment severity	68
Laccase-mediated lignin conversion	Isolated or biomass-associated lignin	Mild oxidative conversion and selective modification of lignin linkages under appropriate conditions	Enzyme cost, reaction time, substrate accessibility, and enzyme stability	Oxidative cleavage or transformation of *β-O-4* and other lignin linkages, with the extent of depolymerization depending on enzyme and reaction conditions	69

Note: The listed effects are qualitative tendencies and depend on biomass source, treatment severity, lignin recovery procedure, purification method, and analytical protocol.

adsorption, hydrogen bonding, dispersion and electrostatic forces, hydrophobic association, covalent bonding, molecular diffusion, and mechanical interlocking. Their contributions depend on substrate composition and morphology, surface preparation, adhesive rheology, application conditions, and curing history.[70,71] No single mechanism applies uniformly to wood, metals, elastomers, and hydrated biological surfaces.

On cellulose- and hemicellulose-rich regions of wood, the phenolic and aliphatic hydroxyl groups of lignin can form hydrogen bonds with polysaccharide hydroxyl groups.[72] When viscosity, surface energy, application time, and pore structure are favorable, the adhesive can penetrate lumens, pits, pores, and surface irregularities. Such penetration increases the effective contact area and promotes mechanical anchoring. Excessive penetration, however, may deplete the bondline and reduce its cohesive integrity. Moreover, because hydrogen bonding is weakened by competitive hydration, wet durability cannot be inferred from initial interfacial adhesion alone. It also requires sufficient network cohesion and controlled hydrophilicity to limit water uptake and interfacial displacement.

The aromatic rings of lignin permit intra- and intermolecular association, including π-π stacking. The extent and preferred geometry of these interactions depend on lignin structure, molar mass, solvent environment, pH, concentration, and aggregation state.[73-75] Aromatic association may contribute to lignin aggregation and cohesive organization, but excessive aggregation can impair solubility, interfacial wetting, dispersion stability, and the accessibility of reactive groups. Its contribution to adhesion is difficult to isolate quantitatively because π-π stacking occurs concurrently with dispersion forces, hydrogen bonding, and hydrophobic association. Figure 2 schematically illustrates possible lignin-polysaccharide interactions in the plant cell wall together with intra- and intermolecular π-π arrangements in lignin.

Covalent network formation uses native lignin functionality or groups introduced by modification. Representative routes include hydroxymethylation followed by phenol-aldehyde condensation,[76] incorporation of epoxidized lignin into amine-cured epoxy networks,[77] urethane formation between lignin hydroxyl groups and isocyanates,[78] and urea-type linkages formed from isocyanates and amino-functional co-reactants.[79] Esterification, etherification, and oxidative coupling offer further routes.[11] These reactions strengthen network cohesion and may form covalent interfacial bonds when complementary substrate groups are available. Excessive functionality or rapid curing, however, can impair wetting, cause premature gelation and internal stress, and increase brittleness. Cure rate, crosslink density, segmental mobility, and energy dissipation must therefore be balanced.

The performance of unmodified technical lignin is often constrained by batch-dependent composition, broad molar-

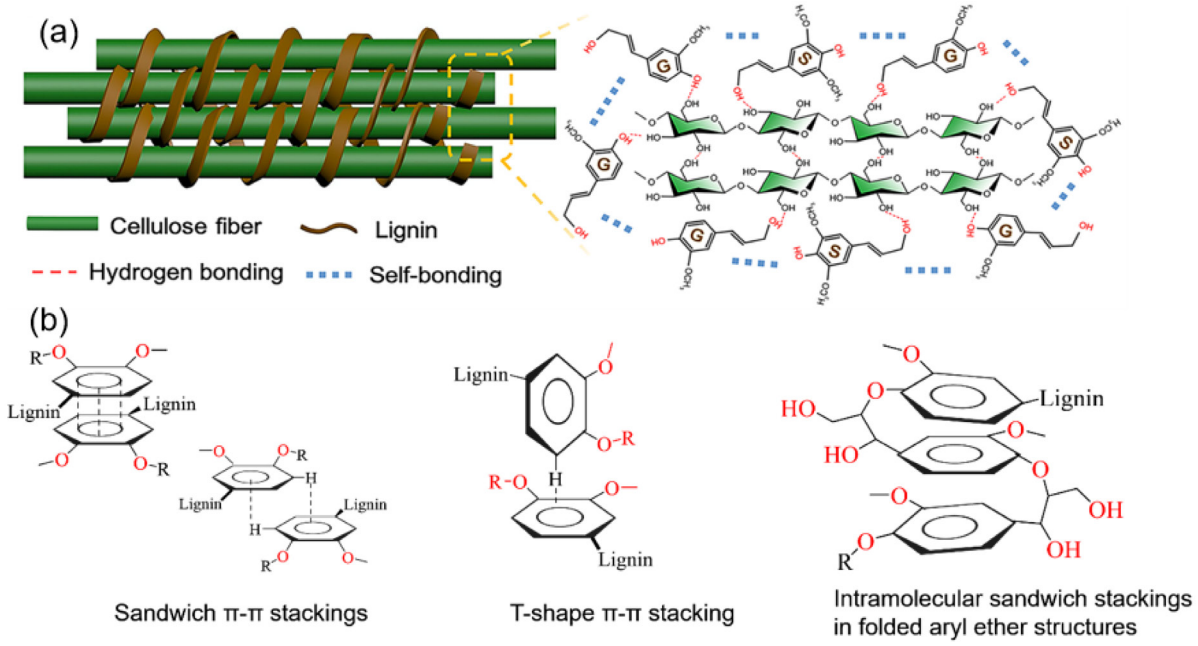

Figure 2. (a) Structural interactions between in situ lignin and cellulose. Reprinted with permission from Ref. 72; (b) Intramolecular sandwich π-π interactions within lignin. Reprinted with permission from Ref. 74.

mass distribution, limited accessibility of reactive aromatic sites, incomplete solubility or dispersion instability, high formulation viscosity, residual impurities, and curing kinetics that are poorly matched to industrial processing cycles.[80-82] These limitations are interdependent and cannot be predicted from total hydroxyl content or a single average molar-mass value alone.

Formulation design therefore requires characterization of each lignin fraction and definition of the processing window relevant to the selected substrate and curing process. Molar-mass distribution, solubility, and solids content influence viscosity, wetting, and penetration; accessible functionality, curing chemistry, and catalyst selection determine reaction rate and conversion; network topology governs cohesive strength, toughness, and stress distribution; and polarity and crosslink density affect water uptake and wet durability. Chemical modification is justified only when it improves these coupled properties without introducing disproportionate reagent hazards, solvent demand, processing complexity, energy consumption, or cost.

Functionalization Strategies and Their Design Consequences

Functionalization is used to adjust reactive-site accessibility, solubility, compatibility, molecular architecture, and curing chemistry.[83,84] Because most functionalization reactions alter several structural variables simultaneously, the reaction name alone cannot predict adhesive performance. Meaningful comparison requires the degree of substitution, accessible functional-group content, molar-mass distribution, residual reagents, solvent demand, product yield, and spatial distribution of the introduced groups to be reported.[80,85-87]

1. Reaction Pathways

Amination introduces amino functionality through reactions such as the Mannich reaction.[88] The chemical role of the resulting aminated lignin depends on whether the introduced groups are primary, secondary, or tertiary amines. Primary and secondary amines can participate directly in epoxy ring opening or react with cyclic carbonates in non-isocyanate polyurethane systems, whereas tertiary amines generally act as curing catalysts or modify lignin solubility and interfacial interactions rather than serve as stoichiometric crosslinking sites. Amination may also induce lignin–lignin coupling or increase molar mass; therefore, nitrogen content alone is insufficient to establish curing functionality.

Epoxidation commonly involves the reaction of lignin hydroxyl groups with epichlorohydrin or other glycidyl reagents to introduce pendant oxirane groups. Epoxidized lignin can function as a reactive component or modifier in amine-cured epoxy networks and can partially replace bisphenol A-derived epoxy resins.[89,90] Its effectiveness depends on epoxy equivalent weight, residual hydroxyl content, molar mass, viscosity, dispersion state, and compatibility with the curing agent. Side reactions between oxirane and residual lignin hydroxyl groups may cause premature crosslinking or reduce the measurable epoxy functionality.

Graft copolymerization introduces polymeric side chains onto lignin through radical polymerization, controlled radical polymerization, or coupling reactions.[91] Graft density, side-chain composition, chain length, and graft distribution govern solubility, matrix compatibility, phase behavior, viscosity, and mechanical properties. Apparent improvements cannot be attributed to grafting alone because ungrafted homopolymer and residual initiator may also affect the reported properties.

Hydroxymethylation introduces hydroxymethyl groups primarily at available aromatic positions under alkaline conditions and can increase lignin reactivity toward subsequent condensation.[92,93] Its effectiveness is limited by the availability of unsubstituted aromatic positions and by competing lignin condensation. Phenolation can cleave selected ether linkages, incorporate additional phenolic structures, increase phenolic hydroxyl content, and expose reactive aromatic sites.[94] The accompanying changes in molar mass and condensation depend strongly on catalyst type, phenol-to-lignin ratio, temperature, and reaction time.

Oxyalkylation reacts lignin hydroxyl groups with alkylene oxides or cyclic carbonates and converts part of the less-accessible phenolic functionality into terminal aliphatic hydroxyl groups. The resulting lignin-derived polyols can exhibit improved fluidity and compatibility with polyurethane components.[95] However, hydroxyl number alone does not define polyol performance; hydroxyl type, functionality distribution, viscosity, residual reagent, and accessibility to isocyanate must also be considered.

These reactions modify several structural variables simultaneously. Their consequences should therefore be evaluated through quantitative functional-group analysis, molar-mass distribution, rheological behavior, cure kinetics and conversion, network characterization, and adhesive

testing under explicitly defined conditions.

2. Structure-Processing-Performance Relationships

Table 2 compares representative lignin functionalization strategies in terms of the resulting structural changes, processing consequences, curing behavior, and reported adhesive performance. Direct comparison among studies is valid only when substrate type and preparation, adhesive formulation, solids content, coating or spreading amount, curing conditions, conditioning procedure, and test method are equivalent. Reported performance values should otherwise be interpreted as formulation-specific results rather than intrinsic properties of a modification method.

Table 2. Representative Lignin Functionalization Strategies and Their Effects on Molecular Structure, Processing, Curing, and Adhesive Performance

Design objective	Modification	Principal structural change	Typical matrix	Functional role	Representative reported outcome	Ref.
Introduction of epoxy functionality	Epoxidation	Introduction of oxirane groups onto lignin	Epoxy adhesive	Enables lignin to participate directly in epoxy curing and partially replace conventional epoxy components	Shear strength of 14.89 MPa and a reported thermal-stability value of approximately 395°C	90
Introduction of reactive functionality	Amination	Introduction of amine-containing functionality into lignin	Soy-protein-based adhesive system	Provides additional reactive functionality and promotes crosslinking within the multicomponent adhesive network	A maximum wet shear strength of 1.07 MPa was reported for the optimized formulation	96
Improved interfacial compatibility	Hydroxyalkylation	Conversion of part of the phenolic hydroxyl functionality into more accessible aliphatic hydroxyl groups	Polyurethane	Improves lignin reactivity toward isocyanate and modifies flexibility and compatibility	Tensile strength of 0.76–4.72 MPa and Young's modulus of 1.85–48.89 MPa were reported for the studied formulations	97
Improved polyurethane processability	Oxyalkylation	Conversion of phenolic hydroxyl groups into terminal aliphatic hydroxyl groups	Polyurethane	Improves polyol reactivity and compatibility; effects on water resistance remain formulation-dependent	A lap-shear strength of approximately 3.1 MPa was reported for the selected lignin-based polyurethane formulation	98
Control of cure chemistry	Amination of fractionated lignin	Introduction of amine functionality into fractionated lignin	Non-isocyanate polyurethane	Provides nucleophilic amine groups for cyclic-carbonate ring opening	Maximum tensile strength of approximately 1.2 MPa and maximum elongation at break of approximately 140% were obtained in different formulations	100
Increased condensation reactivity	Demethylation and hydroxy-methylation	Increased availability of aromatic reactive sites and introduction of methylol functionality	Phenol-formaldehyde resin	Promotes subsequent condensation and network formation in lignin-containing phenolic resins	Adhesive strength increased in the order KLPF < DKLPF < HKLPF under the reported formulation and test conditions	101
Increased reactivity of condensed lignin	Hydrodeoxy-genation followed by methylolation	Modification of condensed lignin followed by introduction of methylol groups at reactive aromatic positions	Lignin-based wood adhesive	Increases accessible sites for network formation and improves lignin reactivity	Improved flowability, water resistance, crosslinking ability, and adhesion were reported relative to the corresponding conventional lignin-based formulation	102

Amination and epoxidation increase the number of groups capable of participating in, or catalyzing, network formation.[88,90,96] Increased reactive-site density can improve cure conversion and cohesive strength but may also shorten pot life, increase viscosity, or cause premature gelation. The effect therefore depends on functional-group type and accessibility, stoichiometric balance, catalyst concentration, and cure schedule rather than on total nitrogen content or epoxy value alone.

Oxyalkylation can increase the proportion and accessibility of aliphatic hydroxyl groups, lower viscosity, and improve compatibility with polyurethane components.[95,97,98] These changes may enhance processability and network homogeneity. However, increased hydroxyl functionality or flexible polyether content can also alter water affinity, crosslink density, and thermal resistance. Improved water resistance should therefore not be presented as a universal consequence of oxyalkylation.

Graft copolymerization can improve matrix compatibility or introduce flexible, rigid, ionic, or stimuli-responsive segments.[75,91,99] Thermal reversibility, self-healing, and shape-memory behavior arise only when the grafted chains

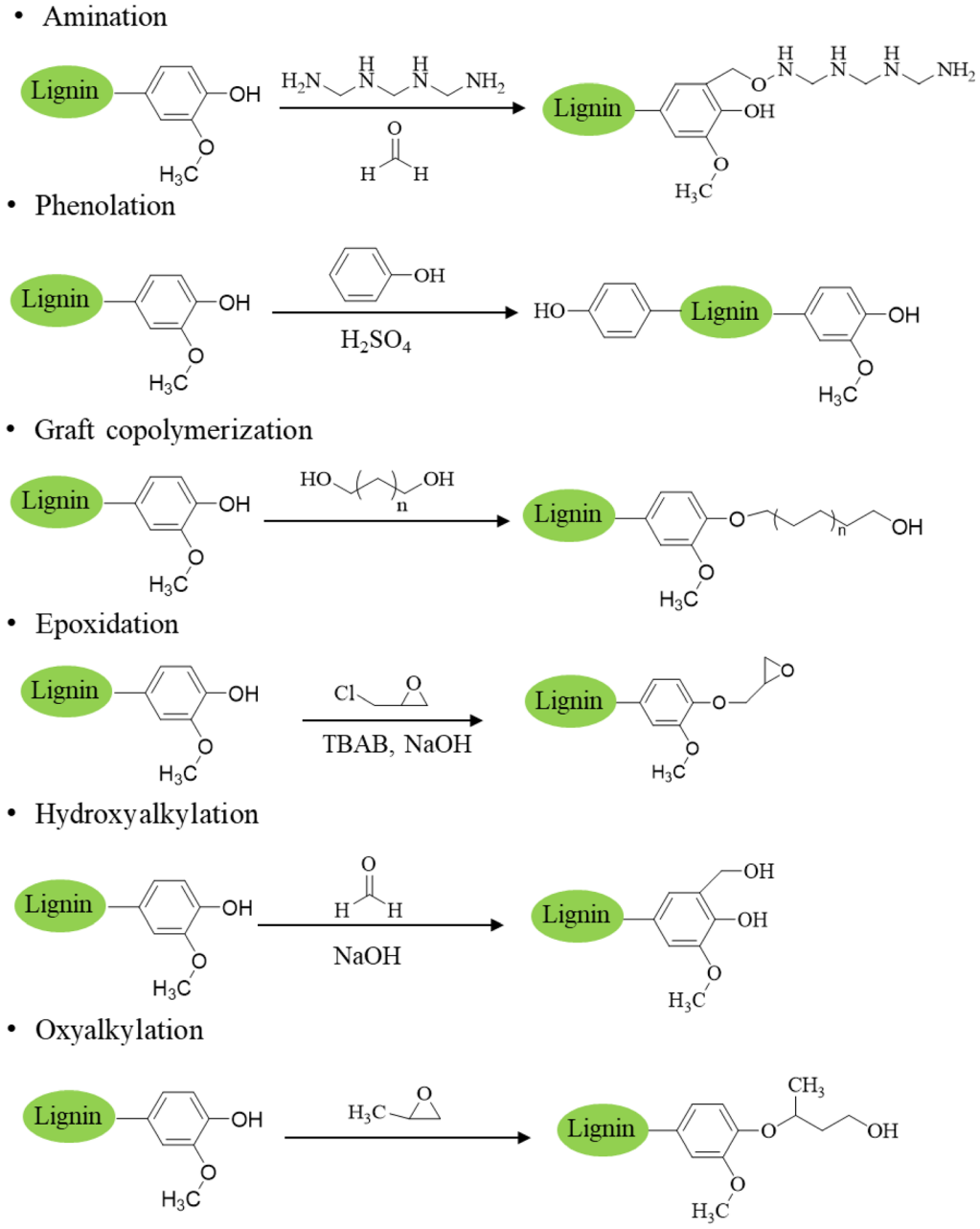

Figure 3. Representative lignin functionalization routes.

or subsequent crosslinks contain appropriate reversible interactions or dynamic covalent bonds; they are not intrinsic consequences of grafting. Moreover, long or densely grafted chains may increase viscosity or produce microphase separation despite improving lignin compatibility at lower graft densities.

The combination of phenolation and amination has been used to modify reactive-site density, flexibility, and solubility,[100] whereas phenolation and hydroxymethylation can increase the availability of reactive sites for subsequent polyurethane formation or phenol-aldehyde condensation.[101,102] These benefits must be weighed against reagent consumption, residual phenol or formaldehyde, purification demand, molar-mass change, and possible condensation during treatment.

Collectively, the reported results demonstrate that adhesive performance is governed not simply by the identity or nominal content of an introduced functional group but by the coupled changes in molar mass, molecular architecture, phase compatibility, rheology, cure kinetics, network topology, and interfacial contact. Functionalization should therefore be selected according to a defined processing and performance requirement rather than treated as an inherently beneficial modification.

Lignin-Based Adhesives: Applications and Performance

Lignin-based adhesives have progressed from partial replacement of conventional wood-resin components to dynamic, weather-resistant, and multifunctional bonding systems. Their performance does not arise from aromaticity alone; it depends on lignin source and isolation history, molar mass and condensation, the accessibility of phenolic and aliphatic hydroxyl groups, residual impurities, compatibility with the surrounding matrix, and the cure and exposure conditions used in each study. To distinguish these roles, this section proceeds from the most direct use of lignin in wood bonding to chemically modified and multicomponent wood adhesives, advanced polymer networks, environmentally durable systems, and specialized interfaces for high-value products.

1. Wood Products and Composites

Wood products remain the most established application platform for lignin-based adhesives because lignin is chemically and physically compatible with lignocellulosic substrates and can contribute to both cohesive interactions within the bondline and interfacial interactions with the cell wall.103,104 Depending on its structural state and formulation, lignin may act as a directly activated binder, a co-component with proteins, tannins, or polysaccharides, or a reactive modifier in a synthetic or bio-based resin network.

1.1 In Situ Lignin Adhesion

Meng et al. examined the organization of in situ lignin in densified bamboo composites and showed that lignin bridges cellulose nanofibrils through hydrogen bonding and physical entanglement (Figure 4a).[72] At an optimum lignin content of 18.3±2.2%, the reported interfacial bonding strength reached 4.4±0.3 MPa. The result demonstrates that lignin already present in the cell wall can become an effective bonding phase when its distribution, mobility, and contact with cellulose are appropriately controlled.

The importance of lignin structure was further clarified by Yang et al., who compared lignins obtained under different isolation conditions (Figure 4b).[105] Less-condensed lignins showed superior bonding because water-induced softening promoted flow and penetration into the porous wood structure, followed by intimate contact and subsequent association or crosslinking with cell-wall lignin. Excessive condensation during isolation restricted molecular mobility and reduced the availability of reactive sites. These findings indicate that isolation severity is not merely an upstream processing variable; it directly governs flow, interfacial contact, and bond development.

1.2 Chemically Modified Lignin in Wood Adhesives

When unmodified lignin lacks sufficient mobility or reactivity for the required cure or service conditions, chemical modification and multicomponent assembly can reinforce the bondline. Wang et al. combined lignin with phytogenic proteins from camellia and soybean meals to produce an aldehyde-free adhesive.[106] Water resistance and antimildew behavior arose from lignin phenolics, protein functionality, and tea saponins. The preservative effect therefore belongs to the complete formulation, which reduced the need for added antimicrobial agents under the reported conditions.

Liu et al. assembled glyoxalated lignin, tannic acid, chitosan, phytic acid, polyethyleneimine, and a triglycidylamine epoxy crosslinker.[107] Glyoxalation increased lignin reactivity, while the other components supplied electrostatic, hydrogen-bond-

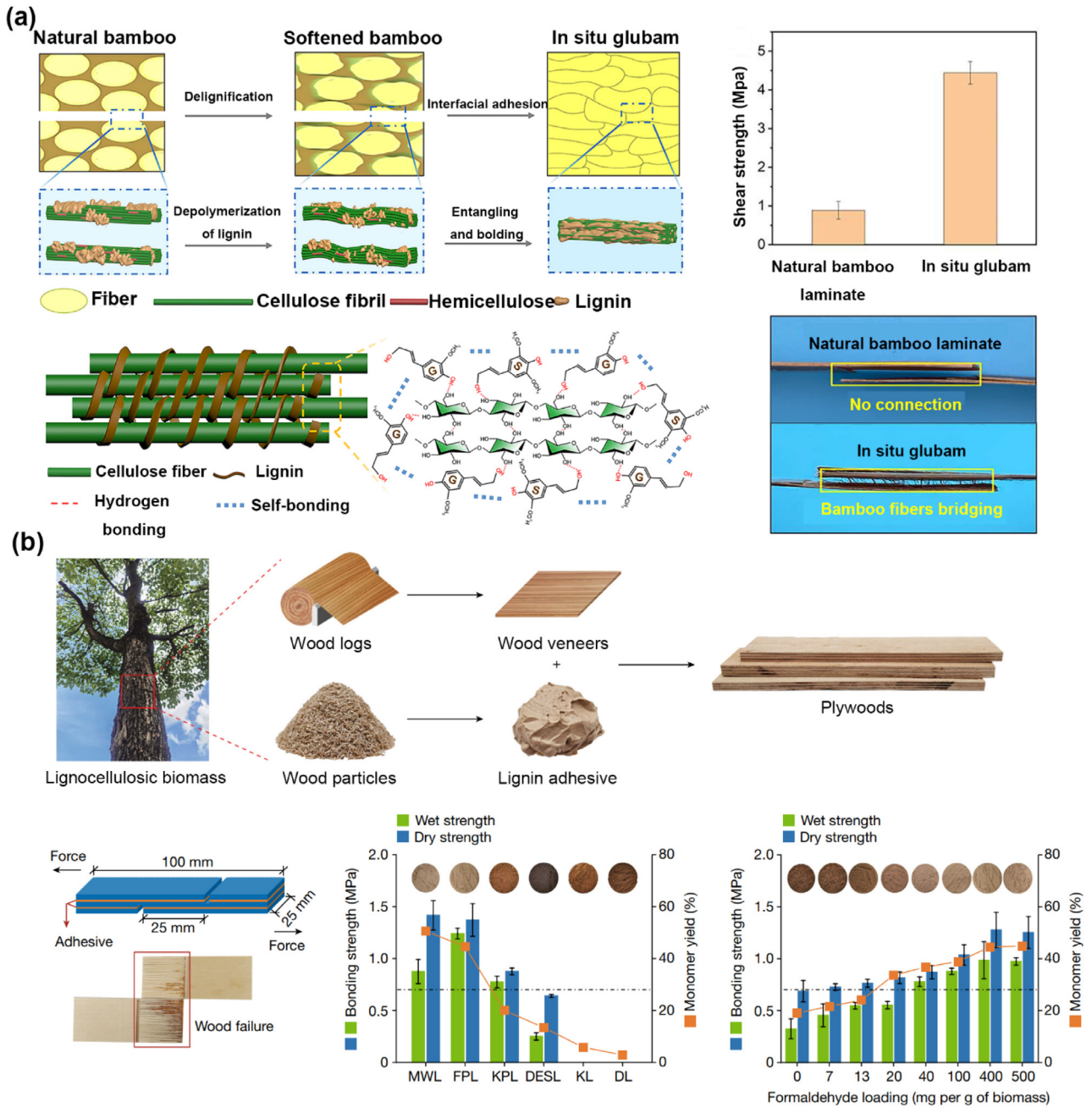

Figure 4. (a) Structural interactions between in situ lignin and cellulose nanofibrils and the corresponding reported interfacial strength. Reprinted with permission from Ref. 72; (b) Effect of lignin isolation and condensation on wood-bonding performance. Reprinted with permission from Ref. 105, respectively.

ing, covalent, and phosphorus-containing flame-retardant functions. The resulting hyperbranched network improved water resistance and flame retardancy, but these properties reflect the complete formulation rather than glyoxalated lignin alone.

Wang et al. combined microwave depolymerization of kraft lignin with phenolic functionalization, then cured the product with epoxidized soybean oil and malic acid.[108] The formulation combined covalent crosslinks, phenolic hydrogen bonding, and exchangeable ester bonds. It reached an aluminum lap-shear strength of about 3.3 MPa and retained 87% of its initial strength after 24 h of water immersion. Controlled depolymerization and dynamic ester exchange thus supported both wet durability and repair.

Effective wood adhesives balance lignin mobility for wetting and penetration, reactive functionality for network formation, and noncovalent interactions for energy dissipation and interfacial stabilization. The optimum balance varies with lignin structure, solids content, stoichiometry, cure schedule, substrate porosity, and exposure.

2. Advanced Lignin-Based Polymer Networks

In advanced networks, lignin can act as a multifunctional crosslinker, a rigid aromatic phase, a source of hydrogen-bond donors and acceptors, and an interfacial coupling component. These roles enable dynamic bonding, repair, and broad substrate compatibility.

Li et al. prepared lignin-based non-isocyanate polyurethanes (LNIPUs) through a one-pot, catalyst- and solvent-free reaction between aminated lignin and a bis(cyclic carbonate) (Figure 5(a)).[109] LNIPU-20% exhibited an aluminum lap-shear strength of 3.09 MPa when cured at 120°C, whereas the strength decreased to 1.13 MPa at 160°C. The adhesive also showed thermally activated bond rearrangement that enabled self-healing, removal, and reuse. Its practical performance therefore depends on the applied thermal history, recovery conditions, and number of reuse cycles.

Zhang et al. combined cyclic carbonate-amine chemistry, polyetheramines, and aldehyde-amine coupling in LNIPU-G adhesives (Figure 5(b)).[110] The networks bonded diverse substrates and resisted water and organic solvents; plywood wet strength reached 1.32 MPa after boiling-water exposure. Polar interfacial interactions, covalent network formation, and flexible polyether segments acted together. Comparisons with commercial polyurethane remain valid only under matched substrate, bondline, cure, and test conditions.

Zheng et al. used a modified lignin polyol and exchangeable oxime-carbamate bonds to prepare a dynamic polyurethane (MLPU; Figure 5(c)).[111] MLPU-2 combined lignin-enhanced microphase separation, dynamic covalent bonding, mechanical anchoring, and substrate-specific

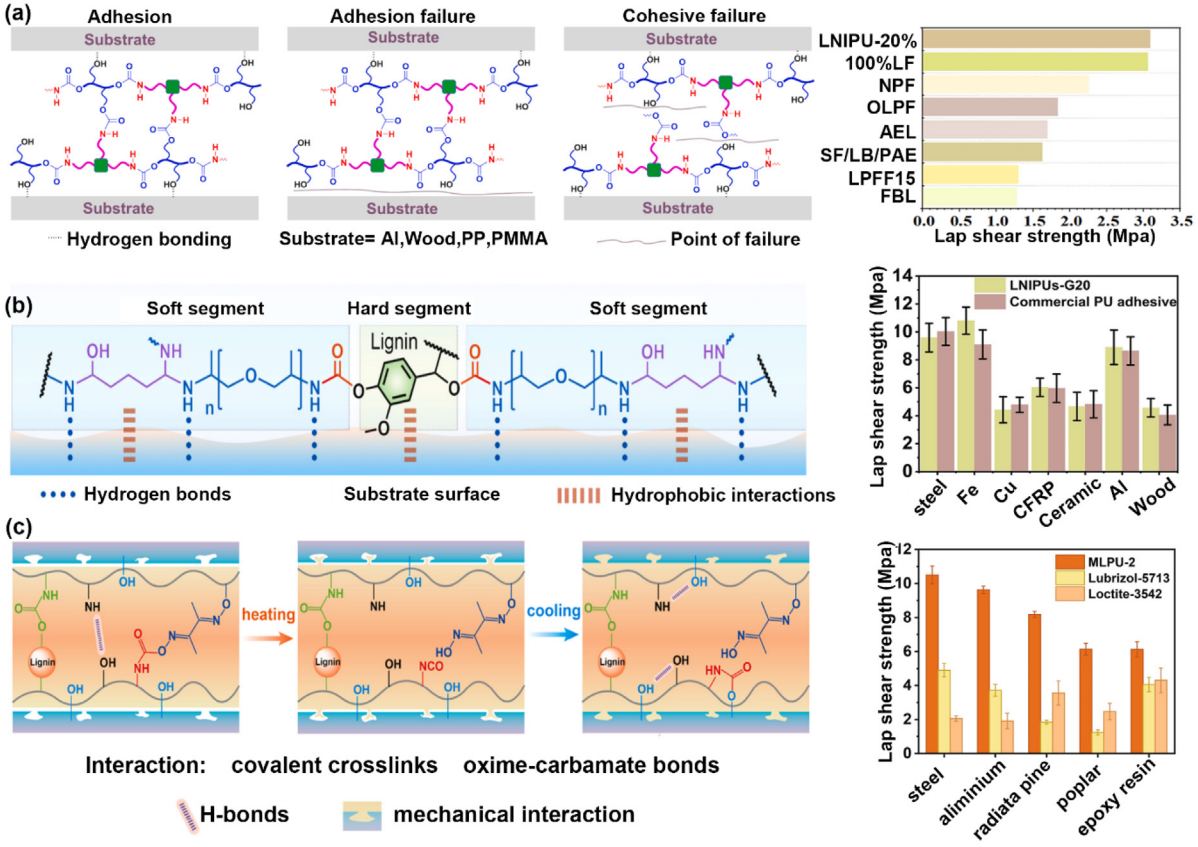

Figure 5. Representative lignin-containing polymer networks for high-performance adhesion: (a) LNIPU formation and bonding mechanism; (b) LNIPU-G network and multi-substrate adhesion; and (c) dynamic covalent MLPU network and lap-shear performance relative to commercial reference adhesives under the conditions reported in the original studies. Panels (a–c) are reprinted or adapted with permission from Refs. 109-111, respectively.

interactions. Under the authors' conditions it outperformed Lubrizol 5713 and Loctite 3542 on several substrates, but the comparison should not be extended beyond the reported processing and test protocol.

Huo et al. prepared formaldehyde-free wood adhesives from waterborne lignin-based epoxy emulsions and polyamide curing agents.[112] Poly(ethylene glycol) chain length controlled fluidity, wetting, penetration, and network flexibility. Fluorescence microscopy directly showed penetration into wood, supporting mechanical interlocking, while spectroscopy was consistent with interfacial hydrogen bonding. This evidence links formulation to interfacial morphology rather than inferring adhesion solely from bulk strength.

3. Environmental Durability and Reversible Adhesion

Adhesives for severe environments must retain cohesion and interfacial contact after thermal cycling, water or solvent exposure, ultraviolet irradiation, high humidity, and chemical attack. Lignin-based systems use aromatic cohesion, dense hydrogen bonding, dynamic covalent exchange, and sometimes photothermal conversion to meet these requirements.

Yang et al. constructed a polyphenol-lignin vitrimer with an exchangeable covalent network (Figure 6(a)).[113] The adhesive combined high cohesion, thermal reprocessability, and repair, while lignin chromophores supplied photothermal conversion. Bonding was retained under the boiling-water, liquid-nitrogen, and organic-solvent conditions tested. Its environmental tolerance arose from dynamic exchange, aromatic reinforcement, and reversible noncovalent interactions.

Li et al. prepared a waterborne polyurethane adhesive from low-molar-mass lignin and carbon dioxide-derived polyols (Figure 6(b)).[114] Adhesion and weatherability were associated with hydrogen-bond density, cohesive-energy balance, nanoscale morphology, and mechanical interlocking. Testing

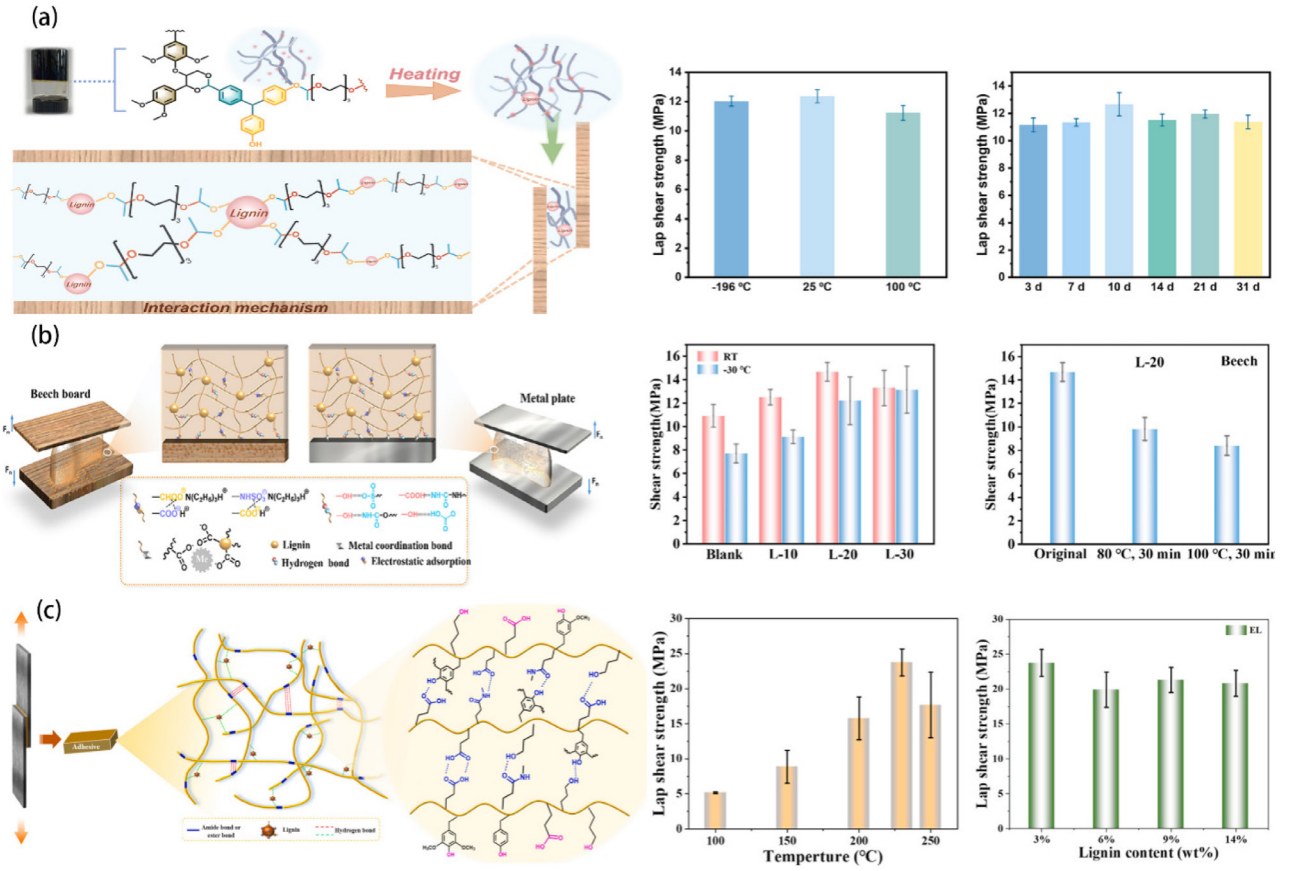

Figure 6. Representative lignin-based adhesive networks designed for severe environments: (a) polyphenol-lignin vitrimer network and environmental resistance; (b) lignin-based waterborne polyurethane bonding mechanism and durability; and (c) lignin/poly(ester amide) network and lap-shear performance under thermal and chemical exposure. Panels (a–c) are reprinted or adapted with permission from Refs. 113-115, respectively.

from −30 to 100°C, at 95%±5% RH relative humidity, and after 72 h of ultraviolet exposure showed how lignin chemistry, soft-segment design, and dispersion morphology jointly control environmental tolerance.

He et al. developed lignin/plant-oil poly(ester amide) adhesives with a dense hydrogen-bond network (Figure 6(c)).[115] Stress dissipation and cohesive integrity supported bonding from −196 to 100°C and after acidic, alkaline, and saturated-salt exposure. Physical and chemical recovery routes were also demonstrated. The breadth of tolerance must nevertheless be read with the exposure time, substrate, conditioning protocol, and failure mode reported in the original study.

4. Specialized and Multifunctional Interfaces

Lignin's aromatic, polyfunctional structure also supports adhesives that combine bonding with ultraviolet shielding, sensing, fatigue resistance, or compatibility with dissimilar high-value substrates. In these systems lignin is one component of a multifunctional interphase, not a stand-alone adhesive.

Li et al. adopted a bio-based dip containing alkali lignin to improve adhesion between polyimide fiber cords and rubber while reducing reliance on resorcinol-formaldehyde-latex chemistry.[116] In this formulation, alkali lignin supplied phenolic hydroxyl functionality for reaction with glyoxal and for hydrogen-bonding interactions with the fiber and rubber matrix, thereby contributing to interfacial anchoring. The treated cords gave an H-pull-out force of 179.6 N versus 178.6 N for the reference, a 180° peel strength of 14.7 N, and fatigue resistance above 67,000 cycles. These results support partial substitution under the reported conditions, not universal replacement by lignin alone.

Wang et al. synthesized lignin-g-P(BA-co-VI) through one-pot reversible addition-fragmentation chain-transfer (RAFT) graft polymerization.[117] Organosolv lignin was first functionalized as a lignin-based macro-RAFT agent and subsequently copolymerized with n-butyl acrylate and vinyl imidazole. The resulting Lignin-BV19.4, containing 19.4 wt% vinyl imidazole, blocked 97.0% of UV-B radiation and exhibited skin-adhesive and self-healing characteristics. Incorporation of LiTFSI introduced ionic conductivity into the compliant graft-copolymer matrix, enabling its use as a strain-sensing material. In this system, the lignin-containing copolymer contributed UV absorption and interfacial adhesion, whereas the grafted polymer chains provided the mechanical compliance required for deformation-dependent sensing.

Zeng et al. incorporated a lignin-nanosphere/MXene-Fe^{3+} system into a conductive hydrogel for wearable sensing.[118] Nanostructuring of kraft lignin increased specific surface area and exposed additional phenolic hydroxyl groups capable of dynamic metal-ligand coordination with MXene-bound Fe^{3+}. The material detected body motion and physiological signals, and polyol addition extended operation to at least −20°C. Because no quantitative measure of adhesion strength was reported, the study supports the use of lignin in robust, mechanically compliant interfaces but does not directly demonstrate improved adhesion.

Across these applications, performance remains sensitive to lignin origin and purification, molar-mass distribution, condensation, hydroxyl accessibility, solids content, crosslinker ratio, cure schedule, substrate preparation, bondline thickness, conditioning, exposure, and failure mode. Reporting these variables is essential for separating interfacial adhesion from bulk cohesion and for assessing toxicity, recyclability, and manufacturing compatibility.

Conclusions and Research Priorities

Lignin is not a compositionally uniform adhesive raw material. Depending on structure and formulation, it can function as a binder, reactive component, crosslinker, reinforcing phase, or functional additive. Its phenolic groups, aromatic backbone, and accessible hydroxyl groups support covalent curing, supramolecular association, and interfacial interaction. Adhesive performance, however, depends on source, isolation history, molar-mass distribution, condensation, functional-group accessibility, residual impurities, and matrix compatibility rather than lignin content alone.

The first priority is to establish reproducible relationships between lignin composition and adhesive performance. Feedstock selection, fractionation, purification, and modification alter molar mass, hydroxyl functionality, solubility, glass-transition behavior, and reactivity. These variables must be optimized with formulation rheology and curing. Higher reactivity or crosslink density may accelerate cure and strengthen the network, but excessive reaction rates or rigidity can narrow the processing window, restrict wetting, promote phase heterogeneity, and embrittle the bondline.

A second priority is to reconcile service durability with

end-of-life function. Dynamic covalent and supramolecular interactions can enable stress relaxation, repair, debonding, or reprocessing, but reversible bonds alone do not establish practical recyclability. Bond-exchange kinetics, activation temperature, repeated repair or reprocessing, retained bond strength, and substrate integrity must be measured. Environmental benefit must also include reagent and solvent demand, separation, recycling efficiency, and compatibility with existing manufacturing.

Industrial translation requires reproducible lignin specifications, storage-stable formulations, application-compatible viscosity, rapid curing, and durability under defined service conditions. Comparisons with commercial adhesives require identical substrates, surface treatments, bondline dimensions, cure schedules, conditioning, exposures, and test methods. Failure mode must distinguish interfacial debonding from cohesive failure. Quantitative relationships among lignin structure, rheology, cure kinetics, network morphology, interface, and long-term performance are needed to separate lignin effects from those of co-resins, crosslinkers, catalysts, fillers, and processing. Toxicity, process economics, energy demand, recyclability, and environmental impact should be compared at equivalent adhesive function and service life.

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