

Robust mussel-inspired adhesives sourced from sustainable lignin

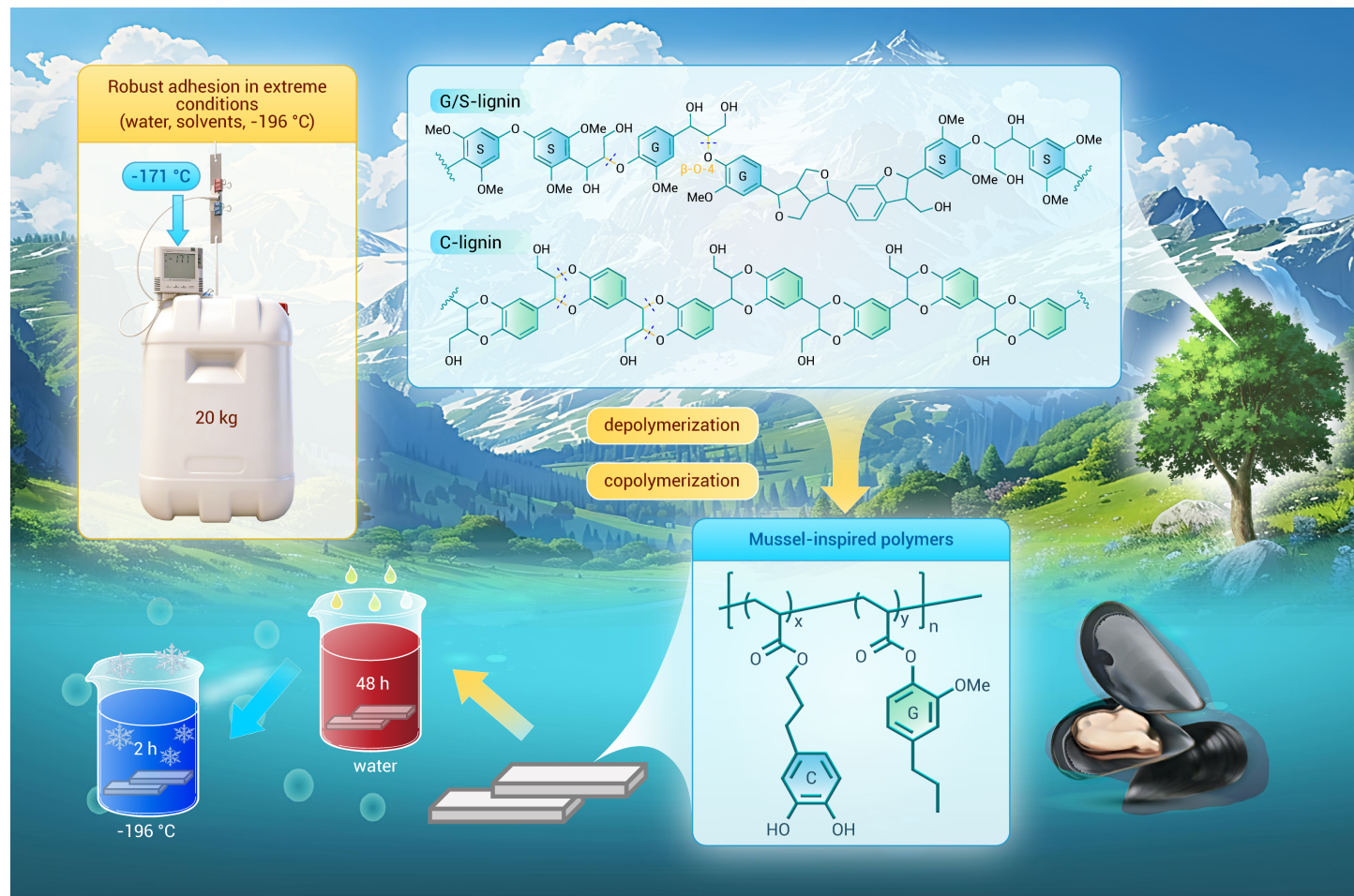
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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- Mussel-inspired adhesives fabricated via a top-down approach from plant lignin.
- Robust adhesion under extreme conditions: water, solvents, and liquid N₂.
- Lignin-specific moieties critically enhance adhesion performance.

Robust mussel-inspired adhesives sourced from sustainable lignin

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Adhesive materials resistant to manifold extreme conditions, such as water, organic solvents, and ultra-low temperatures, are highly sought after but remain rare. Here we present a top-down approach for making mussel-inspired, catechol-based copolymers from two types of natural plant lignins involving depolymerization, derivatization, and copolymerization. The lignin-sourced polymers exhibit robust adhesion and high resistance to water (8.73 MPa) and cryogenic temperature (7.28 MPa), and even retain 6.04 MPa after consecutive immersion in water and liquid N₂, a testing condition rarely examined. Through a combination of monomer permutation screening and molecular dynamics simulations, we demonstrate that lignin-specific moieties, such as catechol, methoxy, and alkyl groups, significantly enhance the adhesive performance of the resulting polymers. Thus, this work pioneers a sustainable pathway for making robust biomimetic materials from bio-based feedstocks.

INTRODUCTION

Adopting biobased feedstocks for producing biomimetic materials is crucial for developing high-performance products while significantly reducing the carbon footprint of the petrochemical industries.^{1,2} Mussel-inspired polymers with pendant catechol units, known for their exceptional underwater adhesion and biocompatibility, are widely used in underwater bonding, coatings, and biomedical adhesives.^{3–5} However, their production largely depends on fossil-derived monomers, requiring harsh conditions and lengthy processes. For example, pyrocatechol, a key precursor for these polymers, is typically synthesized through intermediates like benzene, isopropylbenzene, phenol, and other compounds.⁶ Recent advances in plant catechol-chemistry have led to the development of functional materials with high adhesiveness, mechanical toughness, and antibacterial properties.^{7,8} Despite a mass of mussel-inspired materials exhibiting excellent underwater adhesion, simultaneous tolerance toward cryogenic temperatures and organic solvents remains limited,^{3,4} hindering their broader application in extreme environments such as the poles of the Earth, the outer Planets, and high mountain areas.^{9–11} Conversely, while several adhesive materials have been designed to withstand ultra-low temperatures, their performance in aqueous environments remains a challenge.^{12–14} Therefore, designing a route from sustainable feedstocks for fabricating mussel-inspired polymers with robust adhesion properties and excellent tolerance to manifold extreme conditions remains a critical challenge that demands innovative solutions.

Lignin, a naturally occurring phenylpropanoid polymer, is produced in vast quantities, around 150 billion tons annually through photosynthesis, making it the largest renewable source of aromatic compounds on Earth.^{15,16} Recent advancements have shown that the plant lignin can be efficiently and selectively depolymerized into easily purified aromatic monomers,^{17,18} with product distribution primarily governed by the catalytic systems and lignin's intrinsic structure, thereby unlocking its potential as a valuable feedstock for sustainable chemical and material production. Further studies by Epps III,^{19,20} Sels,²¹ Samec and Wang,²² Barta,²³ as well as our group,^{24,25} have demonstrated that lignin-depolymerized phenolics can serve as sustainable alternatives to fossil-based aromatics in the preparation of well-structured polymers. However, these approaches typically resulted in inert polymers, where lignin's unique groups, such as the phenolic, methoxy, and alkyl groups, have been exhausted or diminished during polymer preparation, leaving them without special properties beyond basic structural functions.²⁶ Targeting functional polymers from lignin, like mussel-inspired materials, aligns well with sustainability goals while enhancing economic potential.

This report presents a novel and sustainable method for preparing mussel-inspired polymers derived from natural lignins (Figure 1). Based on our previ-

ous results, two types of lignin-depolymerized monomers, 4-propanol catechol (POH-C) and 4-propyl guaiacol/syringol (Pr-G/S), were generated from the hydrogenolysis of catechyl lignin (C-lignin) in Chinese Tallow seed coats²⁷ and G/S-lignin in wood sawdust,^{28,29} respectively. These monomers underwent derivatization and copolymerization to produce a series of catechol-functionalized polymeric adhesives. Nearly all atoms from the lignin biopolymers were incorporated into the final materials, demonstrating a high atom economy. The resulting lignin-sourced polymers exhibited robust and reversible adhesion, as well as excellent tolerance towards aqueous, organic solvents, and ultralow temperatures (–196°C). Through a systematic investigation of monomer permutations, we demonstrated that the unique moieties from natural lignin impart enhanced adhesion capacity to catechol-based polymers: (1) the catechol units from C-lignin provide strong interfacial adhesion through hydrogen bonding or coordination interactions, depending on the environmental conditions; (2) the longer propylene linker enhances inter-catechol interactions by facilitating the formation of an interpenetrating polymer network; and (3) the flexible propyl and methoxy groups improves the resistance to ultralow temperature.

MATERIALS AND METHODS

Catalytic hydrogenolysis of C-lignin into POH-C

C-lignin was isolated from Chinese tallow seed coats by following a previous report.²⁷ C-lignin (500 mg), Pd/C (150 mg), and methanol (100 mL) were loaded into a Parr autoclave. The reactor was sealed, purged with nitrogen, and then aerated with H₂ (3 MPa) at room temperature. After the reaction was performed at 220°C for 4 h, the autoclave was cooled to room temperature and depressurized carefully. The lignin oily product was obtained by filtration and subsequent removal of volatiles. Compound POH-C was isolated as a white solid (200 mg) from the oily product by silica gel column chromatography. As calculated, 1.2 g of POH-C was generated from 10 g of Chinese tallow seed coats.

Synthesis of POH-CA

A mixture of POH-C (1.89 g, 11.2 mmol), benzyl bromide (4.61 g, 27.0 mmol), and K₂CO₃ (3.9 g, 33.7 mmol) were refluxed in CH₃CN (30 mL) for 4 h. After cooling, the reaction mixture was filtered, and the solvent was removed under reduced pressure. The crude product was purified by silica gel column chromatography, yielding POH-C-Bn (3.6 g, 92%). Next, to a solution of POH-C-Bn (3.6 g, 10.3 mmol) and Et₃N (1.6 g, 15.8 mmol) in dry CH₂Cl₂ (30 mL), acryloyl chloride (13.2 mmol) was added dropwise at 0°C. The reaction was stirred at room temperature overnight. After solvent evaporation, the product was purified by silica gel column chromatography, yielding POH-CA as a pale yellow oil (4.1 g, 99%).

Production of Pr-G/S from birch RCF

Birch tree chips (20 g, 2–5 mm), RuN/ZnO/C catalyst (2 g), and MeOH (200 mL) were charged into the 300 mL stainless steel reactor equipped with a mechanical stirrer. The reactor was purged with N₂, and then was pressurized with H₂ to 3 MPa. After heating at 240°C for 4 h, the reactor was cooled and depressurized at room temperature. The soluble fraction and solid phase (carbohydrate pulp and catalyst) were separated by filtration with a 0.22 μm filter. Pr-S and Pr-G in the soluble fraction were obtained in pure form (1.25 g and 0.75 g, respectively) by hexane extraction, KOH treatment, and subsequent recrystallization (see Supplemental Information).

Synthesis of Pr-GA/SA

In a typical experiment, a solution of Pr-G (5 g, 30.1 mmol), Et₃N (4.6 g, 45 mmol), and dry CH₂Cl₂ (40 mL) was cooled to 0°C, to which acryloyl chloride

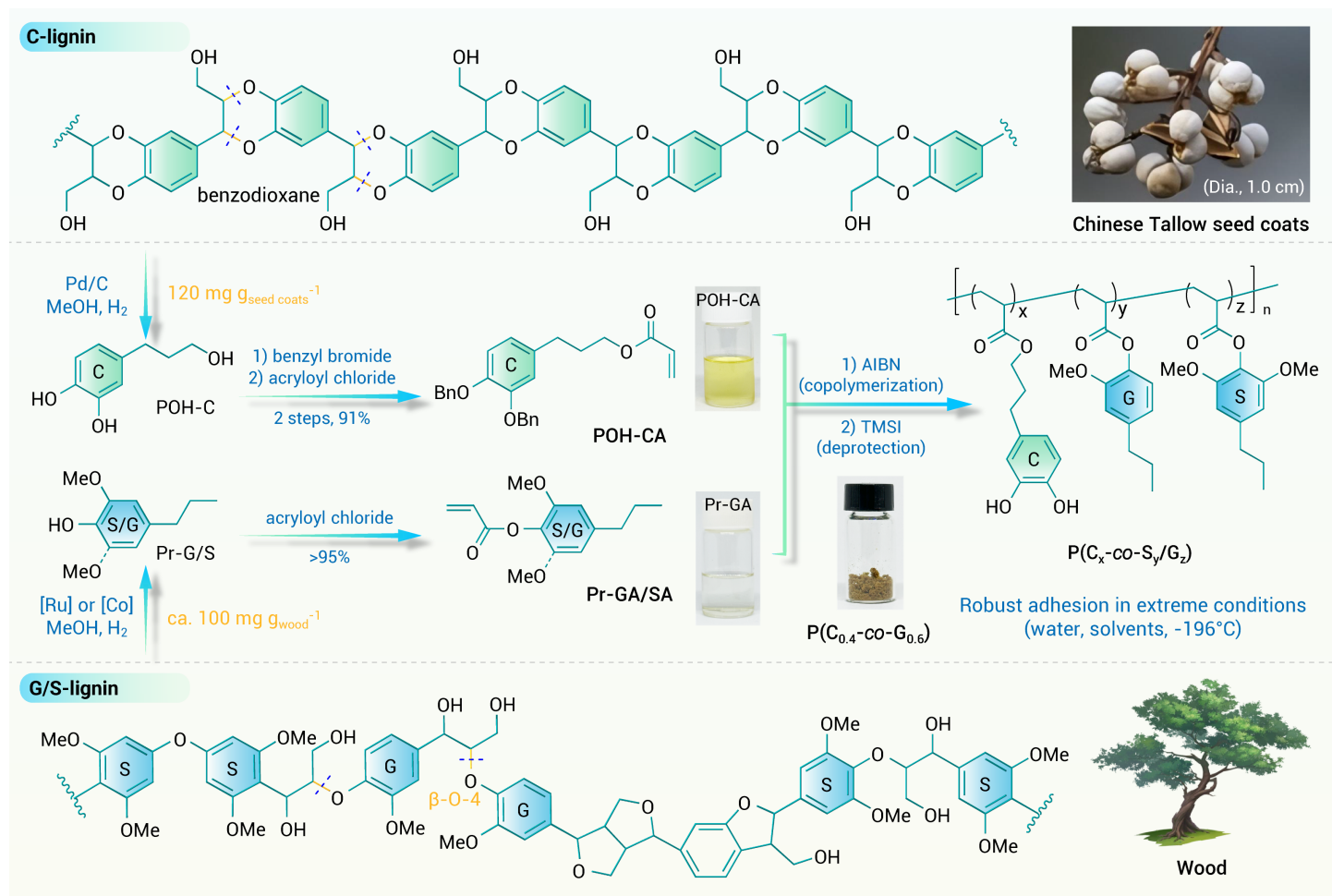


Figure 1. Schematic diagram illustrating design and fabrication of mussel-inspired polymers from lignins.

(5.8 g, 36 mmol) was added dropwise. The mixture was warmed to room temperature and stirred overnight. After solvent removal by a rotary evaporator, the residue was purified by silica gel column chromatography to give **Pr-GA** as a colorless oil (6.3 g, 96%).

Synthesis of P(C_x-co-G_y) copolymers

In a typical experiment, a 10 mL glass thick-walled pressure vessel was charged with **POH-CA** (483 mg, 1.2 mmol), **Pr-GA** (396 mg, 1.8 mmol), azobisisobutyronitrile (AIBN, 15 mg), and DMF (1.5 mL) under N₂.³⁰ After the vessel was heated at 65 °C for 12 h, the mixture was poured into MeOH, from which the copolymer P(C_{0.4}-co-G_{0.6})-Bn was precipitated as a white solid (835 mg, 95%). Next, P(C_{0.4}-co-G_{0.6})-Bn (500 mg) was dissolved in CH₂Cl₂, to which trimethylsilyl iodide (TMSI, 307 mg) was added dropwise at 0 °C. After the mixture was stirred at room temperature for 12 h, HCl aq. solution (1 M) was added. The organic phase was washed with brine and dried over Na₂SO₄. After removing volatiles by rotary evaporator, the residue was washed with hexane, thus affording P(C_{0.4}-co-G_{0.6}) copolymer as a brown solid.

Lap shear test

Steel substrates were mechanically polished and sequentially cleaned with acetone and ethanol. A polymer solution (3 mg in dichloromethane or ethyl acetate) was applied to one substrate (10 × 10 mm), which was then covered with a second substrate and clamped securely. The assembly was cured in a vacuum oven at 100 °C for 1 hour.

The adhered samples were tested in lap shear using a universal testing machine (CMT6103) at a tensile speed of 5 mm·min⁻¹. Measurements were performed on at least five specimens, and the reported values represent the average of the data.

RESULTS AND DISCUSSION

Fabricating mussel-inspired polymers from lignin

Creating mussel-inspired polymers generally requires a catechol-based monomer to provide strong interfacial adhesive properties and a comonomer to fine-tune the polymer's overall properties and functionalities.³⁻⁵ Both types of monomers can be sourced from lignin-depolymerized products, depending on the choice of lignin feedstock and catalytic systems used.^{17,31,32} C-lignin is a homogeneous, linear biopolymer found in the seed coats of certain plants.^{27,33} Its simple composition, comprising simplex caffeyl alcohol subunits and predominant benzodioxane linkages, allows for the selective production of catechol products upon catalytic hydrogenolysis.^{16,32,34,35} We used Pd/C as a catalyst for the hydrogenolysis of C-lignin from Chinese tallow seed coats, which predominantly yielded 4-propanol-functionalized catechol (**POH-C**). Due to its high selectivity (95%), **POH-C** was readily purified in 12% yield based on the weight of the seed coats. Subsequent benzylation of the phenolic groups and the acryloylation of the alkyl hydroxyl group produced a polymerizable catechol-based compound **POH-CA** in high yield (2 steps, 91%).²⁵ In contrast to C-lignin, the more popular G/S-lignin in the hardwood stems primarily comprises sinapyl alcohol (S unit) and coniferyl alcohol (G unit) with β-O-4 as the major linkages.³⁶ The reductive catalytic fractionation (RCF) of birch wood using atomically dispersed Ru²⁹ or Co²⁸ catalysts produced 4-propyl-substituted guaiacol (**Pr-G**) and syringol (**Pr-S**) with a total yield of approximately 100 mg per gram of birch wood. **Pr-G** and **Pr-S** were successfully isolated and purified after treating the RCF mixture through extraction and crystallization.³⁷ Reacting **Pr-G** and **Pr-S** with acryloyl chloride generated polymerizable monomers **Pr-GA** and **Pr-SA** in 96% and 97% yields, respectively (Figure 1).

Following the successful synthesis of lignin-derived monomers, free radical polymerization of **POH-CA** with **Pr-GA/Pr-SA** using AIBN as the initiator,

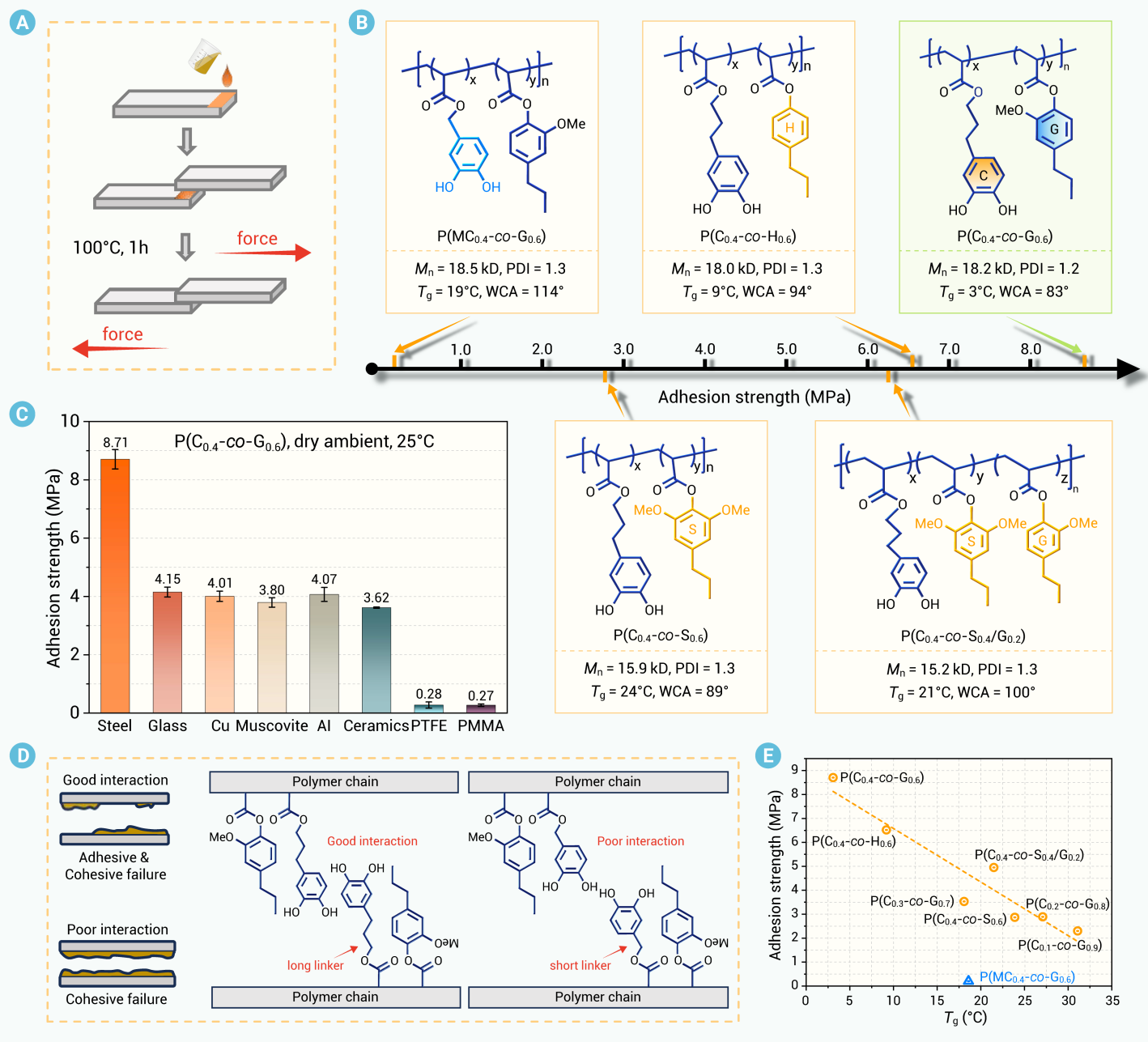


Figure 2. Adhesive properties under ambient conditions (A) Cartoon schematic of the adhesion procedure. (B) Adhesion strength of the resultant polymers on the steel surface (dry, 25 °C). (C) Adhesion strengths of $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ on various surfaces. (D) Comparison of the backbone structure and failure modes of catechol-based polymers. (E) Adhesion strengths of all samples tested as a function of T_g .

followed by TMSI-mediated deprotection of benzyl groups, gave mussel-inspired copolymers $P(C_x\text{-}co\text{-}G_y/S_z)$ as solids in 79–83% yields (Figure 1, see also supplemental information). For comparison, two additional copolymers lacking the lignin-specific moieties were prepared: $P(C_{0.4}\text{-}co\text{-}H_{0.6})$, in which **Pr-GA**/**Pr-SA** was replaced by **Pr-HA** (4-propylphenyl 2-propenoate) lacking methoxy groups, and $P(MC_{0.4}\text{-}co\text{-}G_{0.6})$, featuring a short methylene linker between the catechol and ester units (Figure 2B). Kinetic studies indicated that **POH-CA** and **Pr-GA** exhibit similar reactivity in copolymerization, suggesting a uniform incorporation of C- and G-units along the polymer chain (Table S1). A block polymer, poly($C_{0.4}\text{-}b\text{-}G_{0.6}$), starting from **POH-CA** and **Pr-GA** monomers, was also synthesized. NMR spectroscopies confirmed the structures of these polymers, revealing that the catechol unit ratios in the copolymers were close to the initial feed amounts (see supplemental information). The polymers containing sterically hindered **Pr-SA** moieties exhibited lower average molecular weights (M_n = 15.2–15.9 kD) compared to others (18.0–21.1 kD), while all maintained narrow polydispersity indices

(PDI = 1.2–1.3) (Figure 2B & Table S3). In $P(C_x\text{-}co\text{-}G_y)$ series polymers (where x = 0.1, 0.2, 0.3, and 0.4), increasing catechol moiety content steadily decreased both glass transition temperatures (T_g) and hydrophobicity (Figures S1 & S3). Interestingly, compared to $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ with **Pr-GA** having a single methoxy group, polymers with **Pr-SA** containing two ortho methoxy groups or **Pr-HA** without methoxy groups both exhibited higher T_g and increased hydrophobicity. This likely occurred because the single methoxy in the G unit can increase the flexibility, whereas the two ortho methoxy groups in the S unit restrict flexibility due to steric hindrance.³⁸ Additionally, using a short methylene-linked catechol monomer increased T_g and hydrophobicity, as seen in comparisons between $P(MC_{0.4}\text{-}co\text{-}G_{0.6})$ (T_g = 19 °C, WCA = 114 °) and $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ (T_g = 3 °C, WCA = 83 °). These results suggested that the propylene linker and methoxy group can fine-tune the flexibility and hydrophobicity of the polymers,³⁹ which are essential for optimizing adhesion performance in aqueous and low-temperature environments.⁴⁰

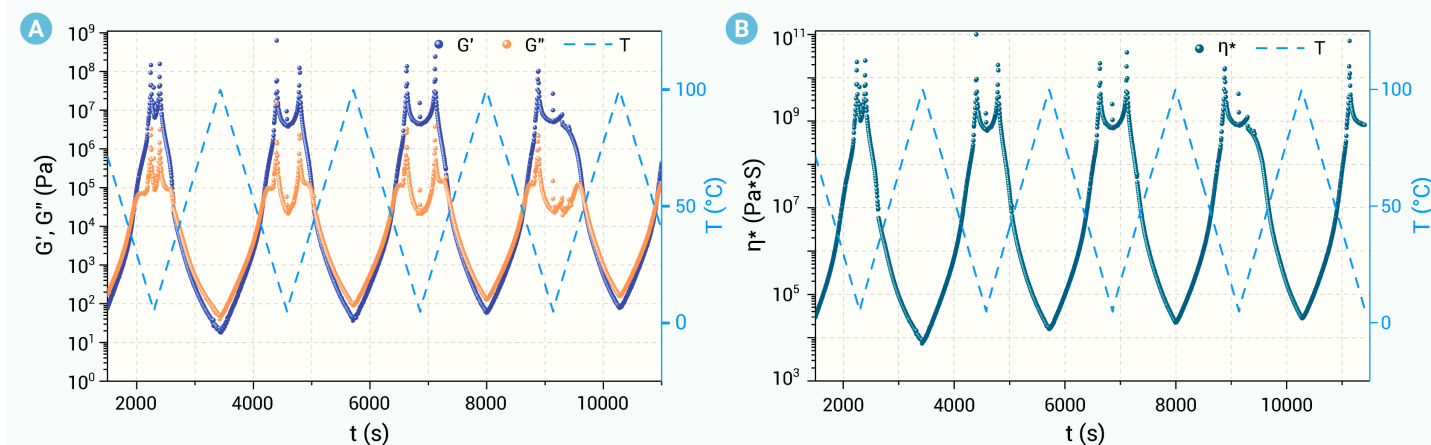


Figure 3. Rheological behaviors of $P(C_{0.4}\text{-co-G}_{0.6})$ during the cooling-warming cycles.

Adhesive properties at dry and room temperature

Introducing metal ions into catechol-based polymers can dramatically enhance adhesive performance by coordinating with adjacent phenolic groups.^{41,42} However, these ions may reduce adhesion and accelerate corrosion over time due to oxidative effects.⁴³ To mitigate these concerns, no metal ions were added in our initial tests. The adhesion strengths of the resulting polymers were initially evaluated under dry conditions at room temperature. The polymer solution was deposited onto steel surfaces and clamped with a binder clip. It was observed that curing temperature significantly influenced the adhesion performance of $P(C_{0.4}\text{-co-G}_{0.6})$, whereas curing time had minimal impact (Table S4). Accordingly, the adhered slides were cured at 100 °C for 1 hour prior to testing (Figure 2A). It was observed that the adhesive performance of these polymers was predominantly governed by the chemical structure rather than by their molecular weight. For $P(C_x\text{-co-G}_y)$ series polymers with similar molecular weights, the increase of catechol content (from $x = 0.1$ to 0.4) led to a substantial increase in the lap-shear strength, reaching a maximum of 8.71 MPa for $P(C_{0.4}\text{-co-G}_{0.6})$ (Table S5). The macroscopic adhesion test demonstrated that the $P(C_{0.4}\text{-co-G}_{0.6})$ -adhered steel slides, with an adhesion area of 2.5 cm², are sufficiently strong to suspend two individuals (~110 kg) (Figure S4). A homopolymer synthesized from **POH-CA** is an elastomer-like material, that is insoluble in all common solvents (such as DMSO, DMF, and CH₂Cl₂) and exhibits no adhesion, likely due to strong hydrogen bonding arising from the abundant and densely packed catechol units. The block polymer, poly($C_{0.4}\text{-b-G}_{0.6}$), showed a low adhesion (0.3 MPa) under such conditions. With a similar catechol content (ca. 40%), replacing the guaiacol unit with the syringol unit, as in $P(C_{0.4}\text{-co-S}_{0.6})$ (2.87 MPa) and $P(C_{0.4}\text{-co-S}_{0.4}/G_{0.2})$ (6.13 MPa), or with a 4-hydroxyphenyl group, as in $P(C_{0.4}\text{-co-H}_{0.6})$ (6.52 MPa), led to a reduction in adhesion strength (Figure 2B). As shown in Figure 2E, the dependence of adhesion strength on T_g suggested that a low T_g is advantageous for strong adhesion in lignin-sourced polymers at room temperature. This is because a higher T_g restricts chain flexibility at room temperature, thereby weakening interfacial interactions and reducing adhesive energy with the substrate. In contrast, polymers with lower T_g exhibit greater chain flexibility, which promotes better substrate wetting and enhances interfacial adhesion.⁴⁴ In the case of polymer $P(MC_{0.4}\text{-co-G}_{0.6})$, in which a short methylene links the catechol to the polymer backbone, a negligible adhesion strength of 0.2 MPa was measured (Figure 2B), albeit with a similar molecular weight to $P(C_{0.4}\text{-co-G}_{0.6})$. The lignin-sourced polymers showed a mixed failure mode, reflecting a well-balanced interplay between interfacial adhesive and cohesive forces.⁴⁵ Conversely, only cohesive failure was observed during the adhesion tests of $P(MC_{0.4}\text{-co-G}_{0.6})$, suggesting that the cohesive strength is lower than the interfacial adhesion (Figure 2D). A plausible explanation is that the longer propylene linker in lignin-sourced polymers distances the polymer backbone and catechol groups,^{13,25,46} thereby facilitating cohesive interactions through the formation of interpenetrating polymer networks (Figure 2D). Several ions, including Fe³⁺, Fe²⁺, and Ca²⁺, were incorporated into $P(C_{0.4}\text{-co-G}_{0.6})$. The resulting catechol-metal

complexes exhibited negligible adhesion, likely due to coordination with metal ions, which strengthens the network through crosslinking and enhances cohesion but weakens interfacial adhesion.^{14,30}

To expand the range of applicable substrates, lap-shear tests were conducted to evaluate the adhesion strengths of $P(C_{0.4}\text{-co-G}_{0.6})$ on various surfaces (Figure 2C & Table S6). The average adhesion strengths on aluminum, copper, muscovite, ceramics, and glass were measured as 4.07, 4.01, 3.80, 3.62, and 4.15 MPa, respectively. Compared with previous supramolecular adhesives, $P(C_{0.4}\text{-co-G}_{0.6})$ exhibited notably strong adhesion, highlighting its promise as a high-performance adhesive system.^{11–15,28} However, it has relatively weak adhesion to hydrophobic surfaces such as polytetrafluoroethylene (PTFE, 0.28 MPa) and poly(methyl methacrylate) (PMMA, 0.27 MPa). This is because, in catechol-based adhesion systems, the primary interfacial adhesive force relies on hydrogen bonding, which is ineffective on hydrophobic surfaces.^{47,48} Nonetheless, the adhesion strengths on PTFE and PMMA remained significantly higher than those of many previously reported supramolecular adhesives.^{45,49}

The viscosity, energy storage, and loss modulus of the optimized $P(C_{0.4}\text{-co-G}_{0.6})$ were further traced by rheology tests (Figures 3A–B). Both the modulus and viscosity of this polymer decreased with increasing temperature. Notably, $P(C_{0.4}\text{-co-G}_{0.6})$ exhibited a transition toward a viscous-dominated viscoelastic state above 45 °C, as indicated by the loss modulus (G'') exceeding the storage modulus (G'), which would be advantageous for interfacial bonding. Furthermore, cyclic rheological measurements revealed the reversibility of G' , G'' , and complex viscosity (η^*) over three cycles, likely due to the reversible noncovalent interactions. However, deviations observed in the 4th cycle may result from partial oxidation of the catechol moieties during rheological tests.

Molecular dynamics (MD) simulations were performed to compare the adhesion behaviors of $P(C_{0.4}\text{-co-G}_{0.6})$ with a long propylene linker and $P(MC_{0.4}\text{-co-G}_{0.6})$ with a short methylene linker (Figure 4).¹² The polymer models, comprising six chains with 72 catechol units and 108 Pr-G units, were used to calculate the cohesive energy density (CED) and interfacial adhesive energy (IAE) with two steel surfaces using Materials Studio.⁵⁰ $P(C_{0.4}\text{-co-G}_{0.6})$ exhibited a CED value of 1.90×10^8 J m⁻³ and an IAE value of -6.79×10^3 kcal mol⁻¹, both higher than those of $P(MC_{0.4}\text{-co-G}_{0.6})$, which had a CED of 1.78×10^8 J m⁻³ and an IAE of -6.58×10^3 kcal mol⁻¹. Under the same shear rate, $P(C_{0.4}\text{-co-G}_{0.6})$ fractured at 654 ps, longer than $P(MC_{0.4}\text{-co-G}_{0.6})$ at 430 ps, indicating $P(C_{0.4}\text{-co-G}_{0.6})$ can sustain a larger shear displacement and exhibits more pronounced spatial-conformational evolution. These differences may arise from the longer, more flexible propylene linker, which strengthens noncovalent interactions and elevates the overall potential energy of $P(C_{0.4}\text{-co-G}_{0.6})$. The cohesive and adhesive energies from MD simulations are governed by the chosen force field, with non-bonded interactions described by van der Waals and electrostatic terms. Both contributions, reflecting π - π stacking and hydrogen bonding, are enhanced under cryogenic conditions (Figure S5). Notably, the simulated shearing behaviors

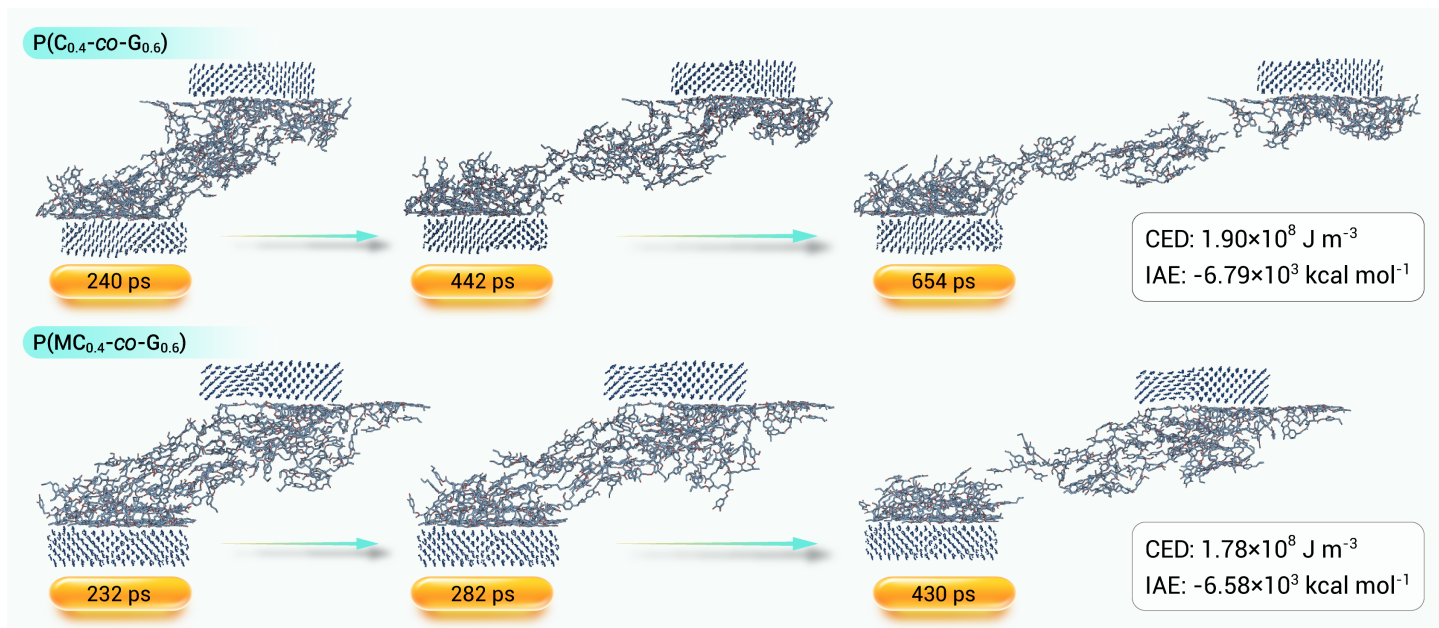


Figure 4. Molecular dynamics simulation of the lap-shear process on steel surfaces.

matched well with macroscopic lap-shear test results.

Resistance to water, solvents, and ultra-low temperatures

Adhesive materials featuring water and low-temperature resistance are crucial for exploration in challenging environmental conditions, such as the Earth's poles, high-altitude regions, and frozen equipment.⁵¹ As is well-known, the catechol-based polymers designed to mimic mussel byssus have exhibited exceptional underwater adhesion, attributed to the diverse interactions facilitated by catechol units with interfacial surfaces (such as hydrogen bonding, coordination, and covalent bonding).^{52–54} Meanwhile, these interactions can enhance the crystallinity of the polymers, which may result in increased brittleness and elevated residual stress as the temperature decreases.⁴⁶ To our knowledge, mussel-inspired adhesives, particularly those not relying on metal coordination,¹⁴ have yet to be applied at ultralow temperatures. Given that the propylene linker, the propyl and methoxy groups sourced from lignin biopolymers endow the hydrophobicity and flexibility of the catechol-based polymers, we hypothesized their potential to withstand water and ultralow temperatures.⁵³

To evaluate the resistance of lignin-sourced adhesives under extreme conditions, the adhered steel slides were subjected to three methods of immersion: in water for 48 hours (25°C), in liquid nitrogen for 2 hours (−196°C), and sequentially in water followed by liquid N₂, before conducting the lap-shear tests (Figure 5A). The average adhesion strengths under extreme conditions, as illustrated in Figure 5C & Table S7, were compared to those measured under dry and ambient conditions. After immersion in water, polymers containing methoxy groups, such as P(C_{0.4}-co-G_{0.6}), P(C_{0.4}-co-S_{0.6}), and P(C_{0.4}-co-S_{0.4}/G_{0.2}), exhibited strengths of 8.73, 2.98, and 5.78 MPa, reflecting slight changes of +0.23%, +0.38%, and −5.7%, respectively, compared to their corresponding ambient adhesion strengths. In contrast, P(C_{0.4}-co-H_{0.6}) without the methoxy group gave a plummeting strength of 2.83 MPa, corresponding to a 56.6% decrease (labeled as brown red), indicating poor water resistance. In terms of direct immersion in liquid N₂, the P(C_{0.4}-co-G_{0.6}) adhesive maintained a high adhesion strength of 7.28 MPa (labeled as lake blue). Furthermore, the P(C_{0.4}-co-G_{0.6}) polymer exhibited no significant contraction or rupture after freezing with liquid N₂, highlighting its excellent flexibility (Figure S6). P(C_{0.4}-co-S_{0.6}) (2.45 MPa) and P(C_{0.4}-co-S_{0.4}/G_{0.2}) (4.8 MPa) also showed approximately 14.6% and 21.7% decrease in adhesion strength, while P(C_{0.4}-co-H_{0.6}) experienced a substantial decline of 63.3% (2.39 MPa). The methoxy group, a distinctive moiety derived from lignin biopolymers, endows the polymers' more resistant ability to water and ultralow temperatures, probably owing to its hydrophobicity, flexibility, and steric hindrance.³⁹ Upon water immersion, hydrophobic and steric domains hinder

water penetration and suppress hydration layer formation at the interface. At ultralow temperatures, the flexible segments accommodate deformation and dissipate thermal stress, maintaining intimate interfacial contact and thereby preserving strong adhesion.

Given the diverse environmental conditions in nature, achieving high adhesive performance at multiple extreme conditions, rather than in isolation, is essential but challenging.^{10,51,55} To this end, we evaluated the shear strength of these polymers after sequentially immersing the adhered steel slides in water and then in liquid N₂, a test rarely performed before. This cascade test poses an unprecedented task and an eye-catching trait because water can infiltrate into polymer matrices, subsequently leading to embrittlement of the materials at low temperatures.^{10,48,54} As shown in Figure 5C (labeled as blue), the adhesive strength of P(C_{0.4}-co-G_{0.6}) experienced a slight decrease, yet remained high at 5.65 MPa after cascade immersion, showcasing its impressive multi-faceted adhesion performance. Other polymers, such as P(C_{0.4}-co-S_{0.6}), P(C_{0.4}-co-S_{0.4}/G_{0.2}), and P(C_{0.4}-co-H_{0.6}) exhibited adhesive strengths of 2.1, 2.2, and 1.87 MPa, respectively. Predictably, P(C_{0.4}-co-H_{0.6}) showed the largest decline of −71%, further confirming the methoxy effect. In a macroscopic test, the P(C_{0.4}-co-G_{0.6})-adhered steel slices can effortlessly support a 20 kg weight after being immersed in water and liquid N₂, and retain their original state upon warming to room temperature, indicating that P(C_{0.4}-co-G_{0.6}) is highly tolerant to water and low temperatures (Figure 5B).

Based on the above results, P(C_{0.4}-co-G_{0.6}) was selected as a sample for subsequent in-depth study (Figure 5D & Table S8). Immersing the steel slices in acidic aqueous solution (pH = 3) and seawater (containing Na⁺, Mg²⁺, and Ca²⁺) did not influence their water resistance adhesions, which resulted in similar shear strengths of 8.02 and 8.65 MPa, respectively. Subsequently, strong adhesion forces were presented as 6.25 and 6.04 MPa after the cascade immersion procedure, which were higher than those from the pure water-liquid N₂ immersion (5.65 MPa). We measured the Raman^{41,42} and Fourier-transform infrared spectroscopy (FT-IR) of P(C_{0.4}-co-G_{0.6}) samples submerged in seawater and liquid nitrogen (Figures 6B & S7). Negligible changes were observed in both cases, further confirming the polymer's excellent tolerance to these extreme conditions. When the steel slices were immersed in an alkaline solution (pH = 11), a slight reduction in adhesion strength was observed, with values of 6.12 MPa after water immersion and 4.54 MPa at subsequent ultra-low temperatures. These decreases are likely due to the reaction of catechol units in the alkaline environment, where quinone coordination predominates interfacial adhesion at high pH, replacing the hydrogen bonding that dominates at low pH.⁵⁴ The P(C_{0.4}-co-G_{0.6}) polymer also exhibited good resistance to some organic solvents. The quantitative adhesion strength remained at 3.70 and 4.30 MPa after soaking in

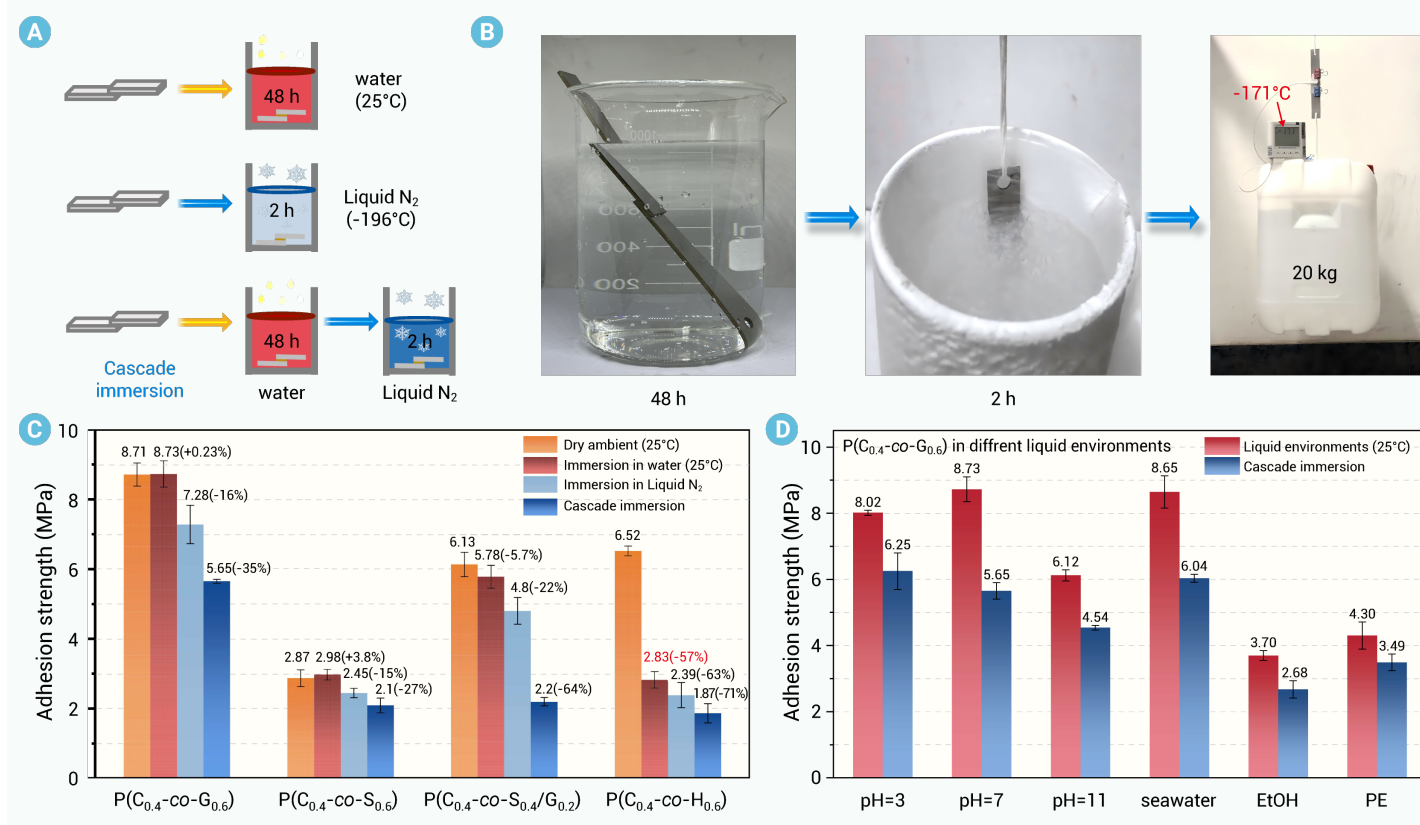


Figure 5. Adhesive performance at multiple extreme conditions (A) Animated processes of immersing the steel slides in water and liquid N₂. (B) The test process of macroscopic adhesion behavior of P(C_{0.4}-co-G_{0.6}). (C) Adhesion strengths of different polymers under different environments. (D) Adhesion strengths under different liquid environments (25 °C) and -196 °C.

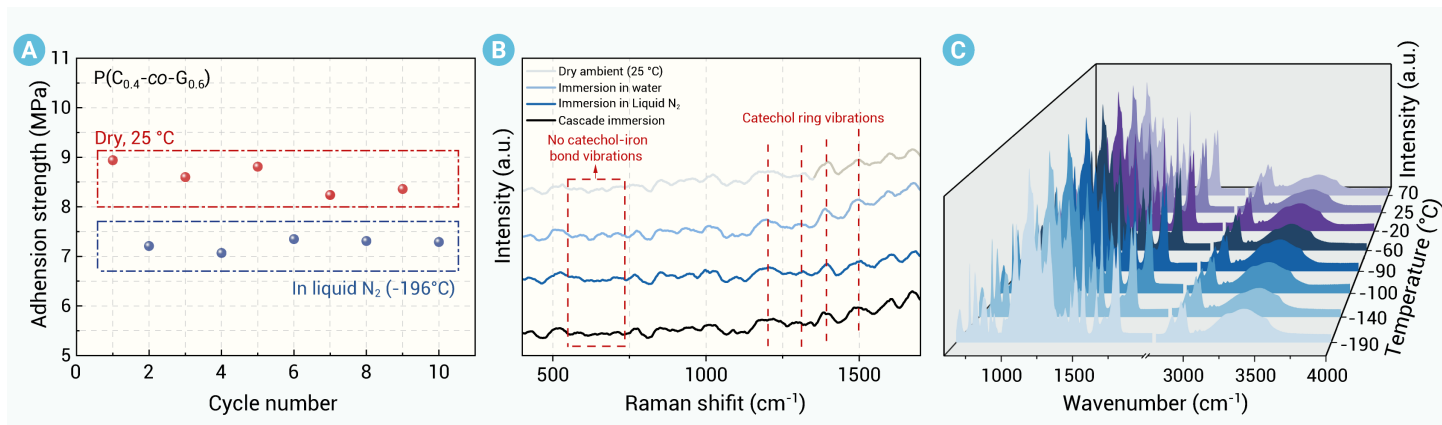


Figure 6. (A) Cycling tests of P(C_{0.4}-co-G_{0.6}) adhesive. (B) Raman spectra of catechol-containing networks. (C) Temperature-dependent FT-IR spectra of P(C_{0.4}-co-G_{0.6}).

ethanol and petroleum ether, respectively. Moreover, cascade immersion in various organic solvents and liquid N₂ resulted in shearing strengths of 2.68 and 3.49 MPa. These results indicated that the lignin-sourced P(C_{0.4}-co-G_{0.6}) adhesive exhibits excellent resistance to ultralow temperatures, water, organic solvents, and even dual treatments, which makes it a highly desirable option for applications in extreme conditions.^{10,51}

We then conducted adhesion-separation recycle tests on steel surfaces, alternating between 25°C and -196°C, up to the 10th recycling (Figure 6A & Table S9). P(C_{0.4}-co-G_{0.6}) demonstrated excellent recyclability, showing no significant degradation in adhesion strength. GPC analysis of the 10th recycled P(C_{0.4}-co-G_{0.6}) polymer manifested no change in average molecular weight (18.0 kD) and molecular weight distribution (PDI = 1.2). These observations suggested that no irreversible chemical transformations occurred during the curing and adhesion processes. The temperature-dependent FT-IR spectra showed negligible changes in the P(C_{0.4}-co-G_{0.6}) polymer signals,

indicating that the interactions within the adhesive remain largely intact over a wide temperature range (Figure 6C).^{51,56}

Based on these results, we utilized the P(C_{0.4}-co-G_{0.6}) polymer to repair a broken agate mortar pestle (Figure 7A).¹¹ After bonding with P(C_{0.4}-co-G_{0.6}) and briefly heating with a heat gun for several minutes, the pestle was able to support a 500 g load, even after ambient water and liquid nitrogen, demonstrating the adhesive's effectiveness in wet and extremely cold environments. Furthermore, by heating an aluminum foil coated with P(C_{0.4}-co-G_{0.6}) for two minutes, we were able to repair an emergency leak in an aluminum can containing ethanol or water (Figure 7B). Remarkably, the adhesive prevented leakage even after exposure to liquid nitrogen, showcasing its resilience in solvent-rich and ultralow-temperature conditions.

Comparison with other adhesives

The established lignin-sourced, catechol-based polymer demonstrated



Figure 7. Illustrations of various applications of $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ adhesive (A) The fractured agate mortar pestle can be repaired and guaranteed its performance after immersing in water and liquid N_2 . (B) The leakage of a barrel holding a solvent (H_2O and EtOH) can be rapidly repaired using a foil paper coated with slightly heated $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ adhesive.

exceptional all-around adhesion performance compared to previously reported adhesives (Table S11). The water-resistant adhesion strength of $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ reached 8.65 MPa, second only to polyphenolic-based copolymers at 10.3 MPa.¹³ Regarding the tolerance to ultra-low temperatures, $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ exhibited an adhesion strength of 7.28 MPa at -196°C , which is superior to most alternatives but still lower than the engineered crown-ether-protein (21.5 MPa)¹⁰ and a nonisocyanate polyurethane (15.9 MPa);⁵⁵ however, these reported adhesives showed reduced or untested water resistant performance. Importantly, $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ maintained high adhesive performance after the cascade immersion in water (or organic solvents) and liquid N_2 , which is otherwise rarely tested.^{14,55,56} These characteristics make $P(C_{0.4}\text{-}co\text{-}G_{0.6})$ a robust adhesive material with tailored applications for rigorous extreme conditions.¹⁰ Notedly, the monomers for synthesizing the current mussel-inspired polymers are sourced from lignin biopolymers, whose inherent functional moieties are almost all retained in the resulting polymers and contribute to their diverse adhesion behaviors. In terms of feedstock, Chinese tallow seed coats are byproducts of biodiesel production, which are rich in C-lignin and capable of yielding the catechol monomer **POH-C**.²⁷ The RCF of birch wood can generate the comonomers **Pr-G** and **Pr-S** while preserving carbohydrates for subsequent valorization.²⁹ This synthetic approach provides a low-carbon feedstock for producing robust mussel-inspired adhesives, especially when considering the entire upstream-to-downstream process. That said, there are several issues with this approach. First, the current derivatization of lignin-derived monomers and polymers relies on classical organic transformations, which involve the sacrifice of certain reagents, such as benzyl bromide and alkaline reagents. Second, large-scale production of catechol and phenolic monomers from lignin hydrogenolysis has yet to be achieved. There is much work ahead for the practical use of lignin-sourced adhesives.

CONCLUSION

In conclusion, we demonstrated a top-down approach to developing robust mussel-inspired adhesives sourced from natural lignins. A key and broadly translatable feature of this work is the generation of catechol and phenolic compounds through the catalytic hydrogenolysis of sustainable C-lignin and G/S-lignin biopolymers. After derivatization and copolymerization, these lignin-derived building blocks enabled the synthesis of a new family of mussel-inspired polymers. The lignin-sourced polymers demonstrated exceptional

adhesion performance, excellent recyclability, and remarkable resistance to water, organic solvents, and ultra-low temperatures, retaining up to 6.25 MPa even after sequential immersion in water and liquid N_2 . Experimental evidence and MD simulations revealed that the exceptional adhesion properties stemmed from the catechol, methoxy, and alkyl groups, distinctive moieties naturally present in natural lignin. Overall, the proof-of-concept demonstrated here offered a pathway to utilize abundant biobased feedstocks for producing functional biomimetic materials, advancing a low-carbon-emission economy beyond existing technologies.

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AUTHOR CONTRIBUTIONS

G.S., S.W., and B.H. conceived and designed the project. B.H. designed and synthesized the adhesives, and performed the adhesives characterizations and lap shear tests. L.W. and F.Z. assisted in molecular dynamics simulation. G.S., S.W. and B.H. wrote the manuscript. All authors discussed the results and commented on the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

Data are available from the corresponding author upon reasonable request.

SUPPLEMENTAL INFORMATION

It can be found online at <https://doi.org/10.59717/j.xinn-mater.2026.100187>.