OPEN ACCESS

Materials Chemistry COMMENTARY

The Innovation Materials

Redox-regulated molecular design towards sustainable hot-melt adhesion

Liwen Yu,[1,2] Yuan Sun,[2] Mengqi Zhu,[2] Wanli Cheng,[2] Zhiguo Li,[2] Yi Lu,[3,4,*] and Siqi Huan[1,2,4,*]

[1]State Key Laboratory of Utilization of Woody Oil Resource, Northeast Forestry University, Harbin 150040, China

[2]Key Laboratory of Bio-based Material Science & Technology (Ministry of Education), Northeast Forestry University, Harbin 150040, China

[3]Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China

[4]Bioproducts Institute, University of British Columbia, 2385 East Mall, Vancouver BC V6T 1Z4, Canada

*Correspondence: siqi.huan@nefu.edu.cn (S.H.); luyi@ipe.ac.cn (Y.L.)

Received: January 7, 2026; Accepted: March 20, 2026; Published Online: March 20, 2026; https://doi.org/10.59717/j.xinn-mater.2026.100211

Citation: Yu L., Sun Y., Zhu M., et al. (2026). Redox-regulated molecular design towards sustainable hot-melt adhesion. *The Innovation Materials* **4**:100211.

Ever since the Stone Age, the pursuit of effective natural adhesives has accompanied the development of toolmaking and engineering activities. Today, however, industrial adhesives, such as urea- and phenol-formaldehyde, heavily rely on petroleum products, which are non-recyclable, non-degradable, and emit toxic by-products (Figure 1A). These human health- and environment-related threats urge the demand for sustainable alternatives.

In response, next-generation sustainable adhesives should be derived from renewable, low-carbon, and biodegradable materials. This includes animal-derived protein- or DNA-based adhesives, biodegradable synthetic polymers, and natural polymer-based adhesives, yet each faces its own limitations. Biomass-derived adhesives struggle with scalability, cost, and storage requirements, whereas biodegradable synthetic polymers may release ecologically hazardous oligomers during degradation.[1] In this context, plant-based polymers, such as cellulose, lignin, hemicellulose, and soy protein, are promising alternatives due to their abundance, renewability, and sustainability.

Further adoption of these bio-based adhesives requires overcoming critical trade-offs among strength, efficiency, and adhesive reversibility. Solvent-based bio-adhesives cure slowly through evaporation,[2] while those utilizing reactive bonding demand energy-intensive crosslinking, with strength susceptible to processing conditions (e.g., temperature, pressure, duration) (Figures 1B_1-B_2).[3] In contrast, hot-melt adhesives (HMAs) offer a promising pathway forward. Their solvent-free, rapid "melt-cool" curing mechanisms provide high efficiency and compatibility in advanced packaging, electronics, and medical applications. A central question arises: how can biopolymers be engineered at a molecular level to satisfy the requirements of meltability, flowability, high strength, and reversible adhesion without resorting to petroleum-based or non-degradable components? The adaptation of bio-based rigid biopolymers to such formats for HMA applications remains a fundamental challenge.

MOLECULAR STRUCTURE DESIGN ENABLING THE MELTABILITY OF BIOPOLYMERS

Converting structurally complex biopolymers into viable HMAs presents significant challenges. Biopolymers like polysaccharides exhibit strong intermolecular interactions that confer high cohesive strength, but these same interactions typically inhibit meltability and flowability, which are essential for thermoplastic applications. Thus, there is a need for dedicated modulation of biopolymers' molecular structures, preferably without introducing petroleum-based or non-degradable components.

Recently, Peng et al. reported a solution using xylan, a hemicellulose, as the first demonstration of bio-based HMAs with reversibility.[4] Unlike cellulose, xylan, as a pentose-based polysaccharide, is not restricted by the presence of a primary hydroxyl group at the C6 position (unlike cellulose), allowing these hemicellulosic molecules to retain their linear backbone while exhibiting greater chain flexibility during strong oxidation treatments. Specifically, $NaIO_4$

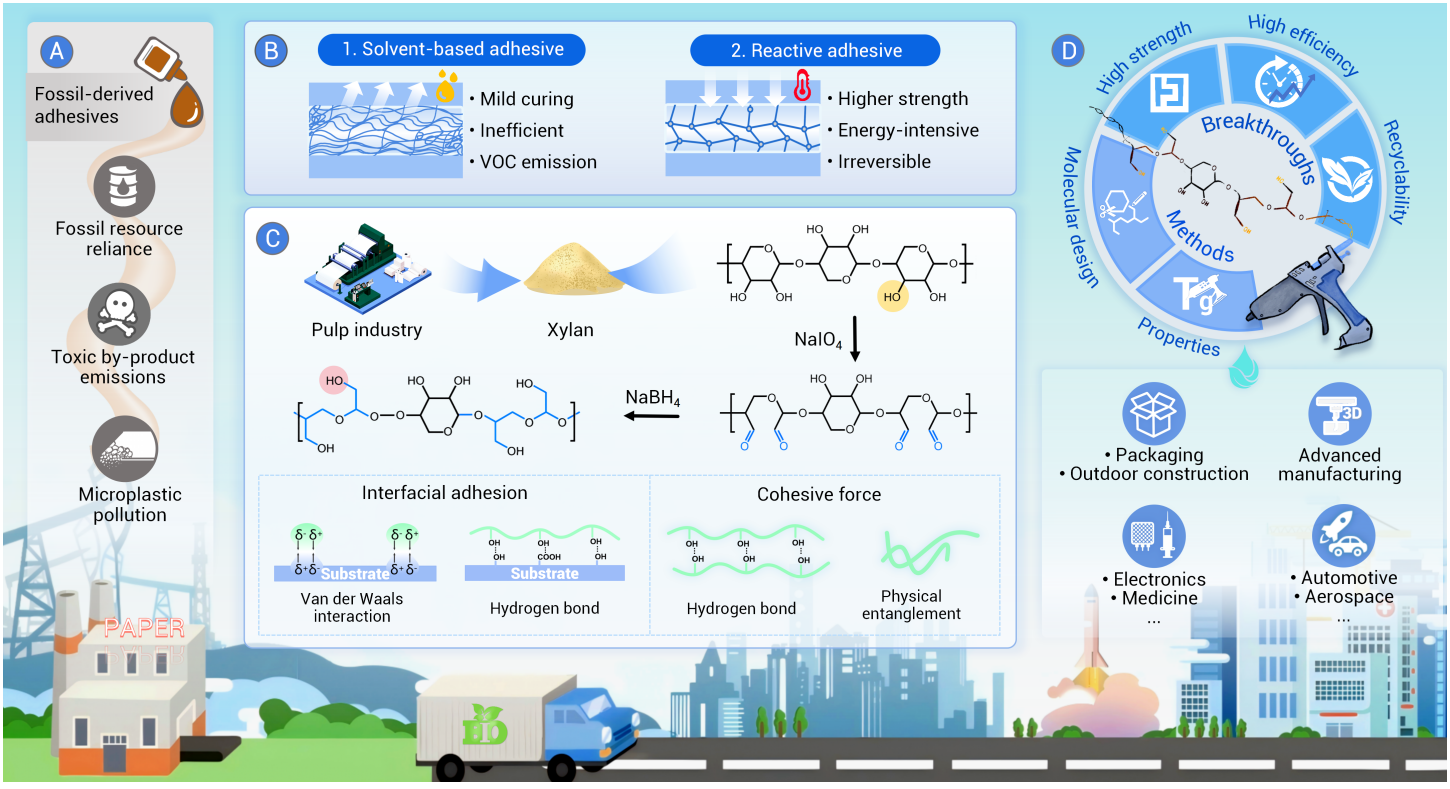

Figure 1. Schematic illustration of the preparation strategy and adhesion mechanism of xylan-based hot-melt adhesives (A) Major challenges associated with conventional fossil-derived adhesives. (B) Schematic representation of the film-forming mechanisms of solvent-based and reactive adhesives. (C) Redox modification of xylan and the resulting interfacial adhesion and cohesive interactions in xylan-based hot-melt adhesive systems. (D) Future development directions and application prospects of natural biopolymer adhesives.

was applied as the oxidant to precisely control the degree of oxidative ring opening at the C2-C3 bond in the anhydroxylose units of xylan, followed by $NaBH_4$ reduction (Figure 1C). This tunable modification of the extracyclic hydroxyl groups lowers xylan's glass transition temperature (T_g) to -13.2–45.9 °C, suitable for hot-melt processing. Computational simulations also confirmed that the redox-induced ring opening shifts the dihedral angles Φ (denoted as C5-O-C1-O) from -66.27° to -130.60°, markedly increasing the rotational freedom around the glycosidic linkage and transforming xylan into reduced dialcohol xylan (RDAX) with extraordinary hot-melt processability.

Based on the experimental observations reported by Peng et al., the thermoplastic processability of hemicellulose can be interpreted from a materials-design perspective. The key to achieving thermoplastic processability in hemicellulose arises from the reversible reconfiguration of intermolecular interactions during heating and cooling, attributed to the successive oxidation-reduction treatments of xylan molecular structures. Importantly, the molecular nature of hemicellulose provides the structural basis for this behavior. Upon heating, hemicellulose's dynamic network preserves segmental mobility, enabling chain softening, spreading, and interfacial wetting within a controlled temperature window, thus fulfilling the thermoplastic flow requirements of HMAs. During cooling, these non-covalent interactions and chain entanglements rapidly reform, restoring high cohesive strength. This places the materials in an optimum regime between structural order and chain flexibility.

These findings demonstrate that precise molecular-level design can transform "difficult-to-melt" biopolymers into programmable HMAs building blocks without compromised reversibility, providing a generalizable design paradigm for bio-based HMAs. Attributed to thermally activated, physically solidified bonding, this approach offers a critical platform for reconciling the longstanding trade-offs among strength, efficiency, and reversibility in bio-based adhesives, establishing a generalizable paradigm for future bio-based HMA design.

BALANCING STRENGTH, EFFICIENCY, REVERSIBILITY, AND RECYCLABILITY

Existing bio-based adhesives typically achieve high bonding strength through high-temperature-induced chemical crosslinking or through physical solidification following solvent evaporation. While such mechanisms promote stable adhesion, they often do this at the expense of reversibility and processing efficiency. In addition, many biopolymers are prone to irreversible chemical transformations, such as Maillard reactions, during high-temperature treatment or repeated thermal cycling, leading to progressive degradation of adhesive performance and further limiting reversibility and recyclability. A common alternative strategy is to achieve reversibility by sacrificing bonding strength, as exemplified by protein-based adhesives that rely on dynamic disulfide bonds.[5]

HMAs enable rapid and reversible bonding via a melt-cooling mechanism, offering a viable pathway to re-establish an integrated balance among bonding strength, efficiency, reversibility, and recyclability in the design of bio-based adhesives. This RDAX adhesive exemplifies the advantages of HMAs, successfully overcoming the traditional trade-off between processability, strength, and reversibility in bio-based adhesives. It combines rapid processing (applied at 105°C and cured within 5-10 minutes) with exceptional mechanical performance, as demonstrated in the reported work by lap-shear strengths of up to 30 MPa on wood substrates and robust adhesion retained at -25°C. Such a performance is not restricted to a narrow processing window or idealized service conditions.

Owing to the deliberate molecular chain design of the biomass polymer, the RDAX adhesive maintains stable bonding strength across repeated bonding-debonding cycles, demonstrating that high performance and reversibility are not mutually exclusive. Such reversible bonding behavior and structural stability are rarely observed in other bio-based adhesives. These results provide direct evidence that high performance and reversibility are not intrinsically antagonistic, thereby offering a solid foundation for the circular utilization of bio-based adhesives.

While the current production cost and carbon footprint may not yet surpass those of conventional ethylene-vinyl acetate copolymer adhesives, the reversible bonding and nondestructive debonding capabilities of bio-based HMAs markedly extend the effective service life of the material, rendering a closed-loop "production-use-regeneration" cycle practically attainable for macromolecular bio-adhesive systems. Ultimately, this xylan-based HMA system advances not by enhancing any single performance metric to its extreme, but rather by molecular-level structural reconfiguration that seeks an optimal balance among strength, efficiency, and reversibility. This paradigm provides a refined blueprint for the molecular architecture of next-generation bio-adhesives.

CONCLUSION AND FUTURE OUTLOOK

Overall, driven by increasing demand for green production and sustainable living, the development of bio-based adhesives has entered a critical turning point and is facing more stringent performance requirements. The study by Peng et al.[4] employs a sequential redox strategy that maintains the molecular weight of natural biopolymers while enabling precise structural and interactive control. Without relying on external additives or irreversible chemical crosslinking, this reported work demonstrates the simultaneous achievement of high bonding strength, rapid processing, and repeatable adhesion, thereby overcoming the long-standing performance trade-offs that have limited the application of purely biomass-based adhesives. Moreover, by introducing a simple recycling and reuse loop, this study establishes a feasible pathway toward natural bio-based adhesive technologies that integrate high performance with circularity.

Nowadays, the evolution of green adhesives is advancing from merely substituting petrochemical feedstocks toward the rational molecular design of pure biopolymers. Future research will increasingly focus on exploiting the intrinsic structures and properties of biopolymers to meet more realistic demands, such as in complex service environments and in high-precision manufactures (Figure 1D). Achieving this will require systematic investigation of how molecular weight distribution, chain flexibility, and hydrogen-bond density influence adhesion strength, moisture tolerance, and thermal reprocessability. Regulating chain flexibility and the spatial distribution of hydrophilic groups can enhance interfacial compatibility and water resistance. Molecular designs that enable repeatable bonding and controllable debonding, together with tunable stimulus-responsive mechanisms, may facilitate selective disassembly and reassembly in electronic and medical devices. Nevertheless, maintaining high strength and long-term stability under harsh conditions (e.g., temperature and humidity fluctuations, cyclic fatigue, and chemical exposure) remains a key challenge for high-end applications, including automotive and aerospace composites. In addition, the fast-curing behavior and hot-melt processability of biopolymer-based adhesives provide opportunities for integration with additive manufacturing and automated assembly. However, understanding shear-dependent rheological behavior, temperature-dependent viscosity stability, and solidification kinetics will be critical for practical implementation.

Natural biopolymer HMAs are expected to play an increasingly important role in renewable materials systems, promoting the transition of the adhesive industry toward low-carbon, circular models. However, a common challenge in converting and utilizing natural biomass is the variability and lack of uniformity in feedstocks, which often lead to fluctuations in product performance and yield. This issue remains a significant barrier to the true industrialization of bio-derived materials, either from plant-based, animal-based, or biosynthetic sources. At present, related formulations and processing routes are still largely confined to the laboratory scale. Achieving controllable molecular construction at an industrial scale, together with systematic evaluation of long-term stability under complex service conditions, remains a critical challenge that must be addressed before practical application can be realized.

REFERENCES

1. Zhao X. L., Wu X. W., Wang Q., et al. (2025). Ecological risks of biodegradable plastics. *Science* **388**:1034–1034. DOI:10.1126/science.adw9060
2. Sun X., Pang Z., Zhu Y., et al. (2023). All-cellulose hydrogel-based adhesive. *Innov. Mater.* **1**:100040. DOI:10.59717/j.xinn-mater.2023.100040
3. Yang G. X., Gong Z. G., Luo X. L., et al. (2023). Bonding wood with uncondensed lignins as adhesives. *Nature* **621**:511–515. DOI:10.1038/s41586-023-06507-5
4. Lv Z. W., Yan X. Q., Jia S. Y., et al. (2025). Bio-based hot-melt adhesive from xylan. *Nat. Sustain.* **8**:827–836. DOI:10.1038/s41893-025-01579-9
5. Lv J., Zhang D. X., Li X. K., et al. (2025). Reversible biobased adhesives enable closed-loop engineered composites. *Nat. Commun.* **16**:7871. DOI:10.1038/s41467-025-62917-1

FUNDING AND ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (32301513). Y.L. acknowledges the "Hundred Talents Program" of the Chinese Academy of Sciences. The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.