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The Innovation Materials

Engineered stereomicrostructures enable tunable biodegradable adhesives

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In a recent study published in Science, Chen and coworkers developed biodegradable adhesives, representing a significant advancement in the field of sustainable polymer materials. This work presents a novel approach through stereomicrostructural engineering to design biodegradable adhesives with tunable properties, offering a promising solution to the environmental challenges posed by petroleum-based adhesives. This groundbreaking research carries significant implications by effectively connecting fundamental polymer science with real-world applications, while providing an eco-friendly substitute for prevailing petroleum-based adhesives in commercial markets.

Adhesives are ubiquitous in modern life, finding applications in electronics, automotive, construction, and packaging industries.[1] However, the vast majority of commercial adhesives currently available on the market are derived from petroleum-based feedstocks. These materials are inherently non-biodegradable and present significant challenges in terms of recyclability, contributing substantially to the global plastic pollution crisis. These adhesives fall into two main categories: thermoset networks, which are cross-linked and non-recyclable, and thermoplastic hot melts, which, while recyclable to some extent, are often not biodegradable (Figures 1A-B). The strong bonds formed by these adhesives with various substrates make their end-of-life management particularly challenging, as they cannot be easily separated and recycled. The environmental impact of these materials has spurred research into more sustainable alternatives.[2] Currently, biodegradable adhesives primarily utilize bio-based materials such as lignin and polysaccharides. However, their performance is still significantly influenced by environmental factors, typically exhibiting bonding strengths of less than 3 MPa. In comparison to petroleum-based products, these adhesives generally demonstrate inferior bonding strength, thereby limiting their practical applications.[3]

Poly(3-hydroxybutyrate) (P3HB), a member of the polyhydroxyalkanoate (PHA) family, has long been recognized as a promising biodegradable and biorenewable alternative to conventional plastics (Figure 1C).[4] P3HB is produced naturally by microorganisms and is biodegradable under both managed (industrial composting) and unmanaged (soil, freshwater, and seawater) conditions. Nonetheless, owing to the intrinsic brittleness of P3HB, successful development of high-adhesion-performance P3HB-based adhesives has rarely been reported in the literature.

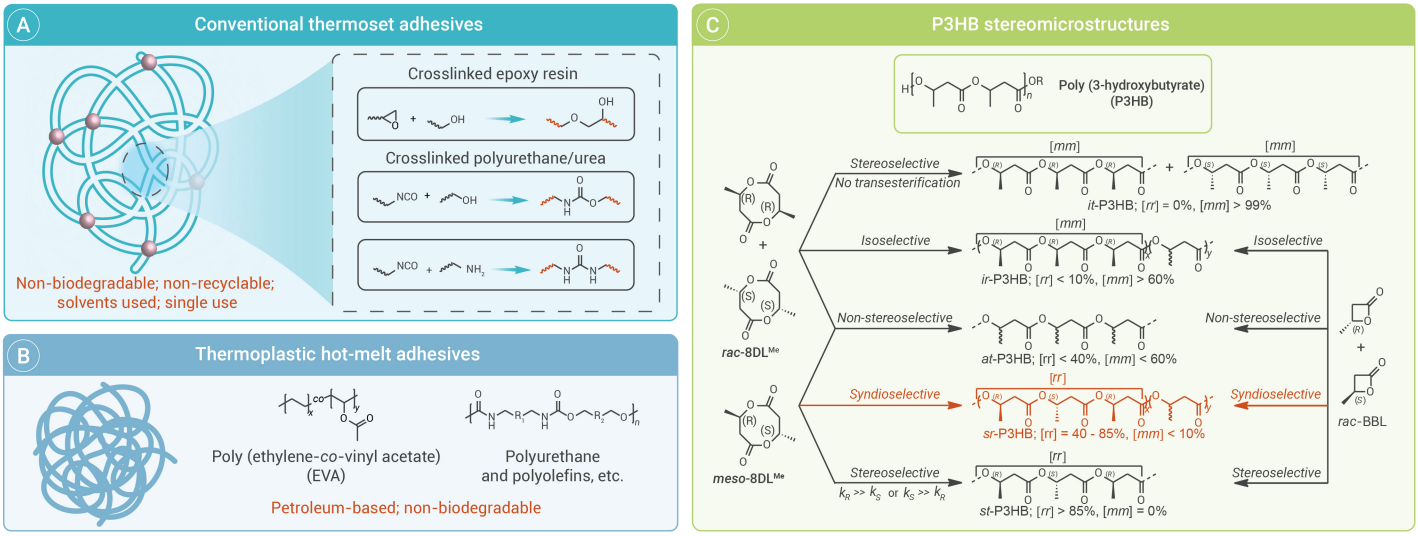

Figure 1. Commercial adhesives and P3HB stereomicrostructures examined in this work (A) Cross-linked network structure of conventional thermoset adhesives with representative motifs (B) Representative semicrystalline polymer structure in thermoplastic hot-melt adhesives, including EVA (C) Stereomicrostructures of P3HB synthesized via catalyzed stereoselective ROP, exploring stereomicrostructure-adhesion relationships. (k_R: ring-opening rate constant at R stereocenter; k_S: ring-opening rate constant at *S* stereocenter).

Recently, Prof. Chen et al. have made a significant breakthrough by elucidating the relationship between the stereomicrostructure of P3HB and its adhesion properties.[5] The authors demonstrated that chemically engineering the stereomicrostructure of P3HB could create materials with superior adhesion strength, outperforming common commercial adhesives. Specifically, they found that syndiotactic-rich P3HB (*sr*-P3HB) exhibited high adhesion strength, while isotactic or atactic P3HB revealed negligible adhesion. The adhesion strength of *sr*-P3HB peaks at 52% [*rr*] triad content (9.5 MPa on aluminum and 10.7 MPa on steel substrates), with significant performance degradation observed at both lower and higher [*rr*] contents, demonstrating a characteristic "low-high-low" parabolic relationship. This discovery is remarkable, as it challenges the conventional view that adhesion properties are primarily determined by chemical composition rather than stereomicrostructure.

The core innovation of this work lies in the stereomicrostructural engineering of P3HB (Figure 1C). Prof. Chen et al. synthesized a comprehensive library of P3HB materials with controlled tacticities, spanning isotactic (*it*-P3HB), isorich (*ir*-P3HB), syndiotactic (*st*-P3HB), and *sr*-P3HB architectures. By precisely modulating the stereochemistry of the polymer backbone, the thermomechanical and viscoelastic properties of P3HB were successfully tailored, which were pivotal for adhesion performance. Notably, the authors identified that the presence of syndiotactic triads ([*rr*] triads) within the poly-

mer backbone was crucial for achieving exceptional adhesion strength. These [*rr*] triads function as physical cross-linking nodes formed a network structure that enhanced energy dissipation and facilitated robust interfacial bonding with diverse substrates. In stark contrast, isotactic P3HB without [*rr*] triads exhibited negligible adhesion. This finding underscored the critical role of stereomicrostructure in governing adhesion behavior, challenging the conventional paradigm that focused solely on chemical composition.

This finding highlighted the critical role of stereomicrostructural control in advanced polymer design. Through precise engineering of P3HB's stereoregularity, the authors achieved materials with finely tuned adhesion properties, paving the way for the development of high-performance biodegradable adhesives that rival or surpass the performance of conventional petroleum-based counterparts.

The adhesion performance of *sr*-P3HB was systematically evaluated through lap-shear adhesion tests on various substrates, including aluminum, steel, glass, and wood. Notably, *sr*-P3HB exhibited adhesion strengths ranging from 1.9 MPa to 9.5 MPa, depending on the specific stereoregularity of the polymer. These values not only exceeded those of common commercial adhesives, such as Gorilla Glue and J-B Weld, but also surpassed most biodegradable bio-based adhesives. Moreover, the adhesion strength of *sr*-P3HB was found to be largely insensitive to molar mass and reprocessing, making it highly suitable for large-scale production and reuse. It was demonstrated that *sr*-P3HB could be repeatedly melted and reprocessed without significant loss of adhesion performance, a feature that is not achievable with traditional thermoset adhesives. This reprocessability, combined with the material's biodegradability, positions *sr*-P3HB as a truly sustainable alternative to conventional adhesives. Remarkably, *sr*-P3HB maintains stable adhesion performance across a wide temperature range (< 245°C) and under high humidity (77% RH) due to its tunable [*rr*] triad-controlled crystallization.

To assess the feasibility of large-scale adoption of *sr*-P3HB adhesives, a detailed techno-economic analysis (TEA) and life cycle assessment (LCA) were performed. The TEA revealed that the minimum selling price (MSP) of *sr*-P3HB was slightly higher than that of ethylene-vinyl acetate (EVA), a commonly used thermoplastic hot-melt adhesive. However, this price gap is expected to narrow with further optimization of production processes and the implementation of more cost-effective catalysts. The LCA highlighted that the current environmental footprint of *sr*-P3HB production is greater than that of EVA, primarily due to the dependence on fossil-derived feedstocks, such as propylene oxide (PPO) and carbon monoxide (CO). Nonetheless, transitioning to bio-based feedstocks and improving polymerization efficiency could significantly reduce this impact. Additionally, the inherent reprocessability of *sr*-P3HB offers opportunities for direct reuse, providing further avenues to lower its environmental footprint and enhancing its potential as a sustainable adhesive solution.

The practical applicability of *sr*-P3HB was demonstrated through a series of real-world experiments, showcasing its potential as a sustainable alternative to conventional materials. By molding *sr*-P3HB into glue sticks compatible with standard commercial glue guns, the material proved to be a viable direct replacement for EVA. In comparative adhesion tests, *sr*-P3HB outperformed EVA, exhibiting higher adhesion strength and successfully supporting greater loads without failure. These results highlight *sr*-P3HB's potential as a high-performance, eco-friendly alternative to traditional hot-melt adhesives, with promising applications across industries such as packaging, construction, and more.

The groundbreaking work by Chen et al. have made a transformative advancement in the development of sustainable adhesives. Through innovative stereomicrostructural engineering, a biodegradable adhesive has been engineered, exhibiting performance metrics that not only rival but also, in certain aspects, surpassed those of traditional petroleum-based adhesives. This research delivered a compelling solution to the global plastic waste crisis while simultaneously unlocking new possibilities for the design of sustainable polymers with precisely tailored properties. Future work should focus on optimizing production processes for cost efficiency and reduced environmental impact. The integration of advanced catalytic systems and the utilization of renewable bio-based feedstocks could further bolster the competitiveness of *sr*-P3HB adhesives against conventional materials. Moreover, expanding the exploration of *sr*-P3HB into diverse applications, such as functional coatings and high-performance composites, holds significant promise for broadening its technological and industrial impact.

In summary, this work presented P3HB by Chen et al. stands as a seminal contribution to polymer science, unequivocally demonstrating that sustainable materials can achieve the performance benchmarks set by their petroleum-derived counterparts. This work bridges critical gaps in the fundamental understanding of polymer microstructure-property relationships, offering a strategic pathway toward sustainable material design and application in materials science.

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AUTHOR CONTRIBUTIONS

S.-L. Zou wrote the manuscript. L.-P. Xiao and R.-C. Sun reviewed and commented on the manuscript. All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.