Polymer upcycling

https://doi.org/10.1038/s44160-025-00928-y

Electrochemical upcycling of polyolefins into adaptable networks

Rhys W. Hughes & Brent S. Sumerlin

Electrolysis enables selective dual C–H functionalization for transforming inert polyolefin deconstruction products into dynamic recyclable networks.

Thermoset plastics are difficult to recycle due to their crosslinked structure. This challenge becomes especially problematic for carbon fibre-reinforced polymer (CFRP) composites valued in aerospace, automotive, and renewable energy applications for their strength and low weight[1]. Although recent advances have enabled the triggered deconstruction of these composites to recover carbon fibres, the process often yields a substantial fraction of low-molecular-weight oligoolefins[2]. These fragments are chemically inert and structurally complex which complicates approaches to upcycling these materials or recovering the original feedstock.

Now, writing in *Nature Synthesis*, Moore and co-workers report an innovative electrochemical strategy that installs distinct functional groups onto polyolefin oligomers[3]. A cooperative electrolysis enables direct C–H functionalization under mild conditions to convert waste fragments into covalent adaptable networks (CANs), a recyclable class of thermosets (Fig. 1). This elegant electrochemical strategy offers a promising route towards a circular polymer economy, where plastic waste from composites can be transformed rather than discarded.

Direct C–H bond activation is challenging, especially for complex branched oligomers such as those derived from dicyclopentadiene (DCPD)-based thermosets. Traditional C–H functionalization methods often require high temperatures (≥120 °C) and use harsh reagents that may trigger uncontrolled crosslinking or degradation. Electrochemically driven macromolecular transformations are still relatively rare[4–7], yet they offer some advantages[8,9], such as milder conditions, more selective activation[10], and compatibility with renewable electricity. The work by Moore and colleagues exemplifies how this approach can unlock reactivity in complex systems.

A mediated electrolysis strategy is successfully demonstrated under ambient conditions. *N*-Hydroxyphthalimide (NHPI) is oxidized at the anode to form a phthalimide-*N*-oxyl (PINO) radical[11], which selectively abstracts hydrogen atoms from allylic C–H bonds on the polymer backbone. The resulting polymer radicals are intercepted by *tert*-butyl hydroperoxide (*t*-BuOOH), a co-oxidant that installs ketone functionalities while suppressing radical recombination. Simultaneously, some phthalimide groups are also attached to the polymer, forming a nitrogen-based functionality. The result is a one-step dual functionalization, where a carbon–oxygen bond (via ketone formation) and a carbon–nitrogen bond (via phthalimide) are formed. This combination has not previously been achieved in a single operation, and it may pave the way for dynamic covalent chemistry on otherwise inert polymer chains, exemplifying how electrochemical approaches can unlock reactivity in complex systems.

Another interesting feature of the demonstrated approach is its selectivity. Tertiary allylic C–H bonds located along the polymer backbone are preferentially activated over secondary allylic positions. This outcome is confirmed through direct competition experiments using small-molecule analogues. Such control is crucial and ensures that functional groups are introduced at well-defined positions, helping to avoid excessive crosslinking or degradation. Additionally, the degree of functionalization can be tuned by adjusting electrolysis time or applied potential. This level of programmability is promising for future scale-up and tailoring of material properties.

These modified oligomers can then be transformed into new materials. After electrolysis, phthalimide groups are converted into amines using an Ing–Manske procedure. Following amine-deprotection, dynamic covalent imine and oxime bonds are formed with the ketone units. This crosslinking yields a CAN that behaves like a traditional thermoset but can also be reprocessed when exposed to elevated

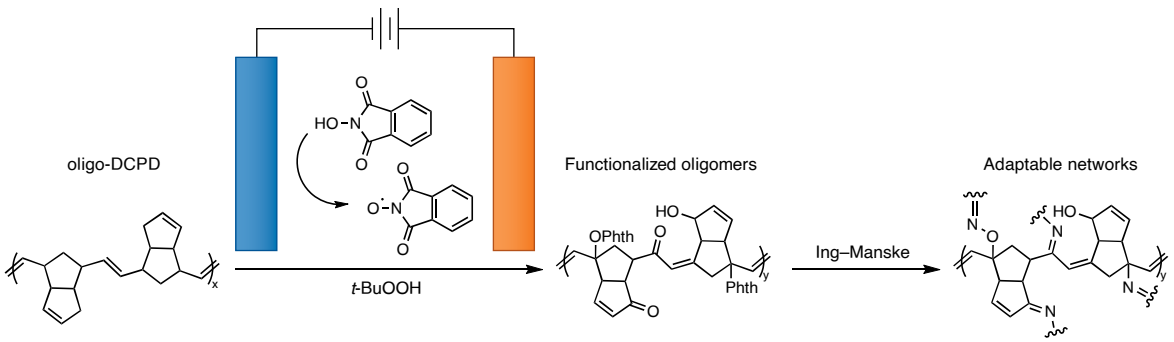

Fig. 1 | Electrochemical upcycling of polydicyclopentadiene oligomers via C-H activation. The first reaction step represents the electrochemical functionalization of dicyclopentadiene oligomers via C–H activation to install ketone and protected amines. The following Ing–Manske deprotection step reveals amines that can react dynamically with the installed ketones.

temperatures. Thermal analysis reveals that films cast from these materials retain elasticity and mechanical integrity above 150 °C, while films made from unmodified oligomers crack or melt. Critically, the CANs resist dissolution in solvent unless treated with an excess of a small-molecule amine, which breaks the dynamic bonds. On-demand deconstruction therefore allows for multiple use cycles. After breaking the network, the oligomers can be re-functionalized or repurposed, achieving a degree of recyclability previously unattainable for most thermosets. The process is also found to be scalable to decagram quantities, and the strategy can be extended to other polyolefins, including rubber-like polybutadiene, expanding its impact beyond DCPD-based composites.

The reported electrochemical advance complements a growing array of strategies for designing sustainable thermosets, including dynamic covalent networks and biobased formulations. By addressing post-use upcycling of waste polymer fragments, this work helps to resolve a key challenge in the circular lifecycle of composite materials. There are, however, opportunities to continue advancing the demonstrated method. The inherent heterogeneity of oligomer mixtures poses challenges for uniform functionalization across different chain architectures. Encouragingly, a growing library of redox mediators and advances in electrochemical C–H activation offers promising tools for this task.

It will also be important to extend this approach beyond allylic C–H sites to fully saturated backbones, improve control over functional group distribution, and minimize reliance on stoichiometric oxidants for practical implementation. Electrochemistry is clearly emerging as a powerful tool for polymer scientists interested in polymer deconstruction and creative redesign. This approach challenges the community to rethink waste streams as chemical feedstocks and to integrate electrical inputs into polymer processing and recycling strategies. The success of electrochemically driven functionalization may inspire new strategies for adapting inert polymer waste into value-added, upcycled materials.

Going forward, the development of new mediators and electrochemical reaction cell designs could unlock even more selective or efficient modifications, paving the way towards scalable upcycling of a wider range of plastics. If successful, this direction could establish electrochemical functionalization as a cornerstone of sustainable polymer chemistry.

By combining radical chemistry with electrochemistry, Moore and co-workers report a promising route for transforming plastic waste into dynamic, high-performance materials. The dual C–H functionalization strategy achieves a longstanding synthetic goal: the conversion of low-value polyolefin fragments into recyclable thermosets under mild, tunable conditions. This work sets a new benchmark in polymer functionalization while illustrating the broader promise of electricity-driven manufacturing in materials science.

Rhys W. Hughes & Brent S. Sumerlin ✉
George and Josephine Butler Polymer Research Laboratory, Center for Macromolecular Science and Engineering, Department of Chemistry, University of Florida, Gainesville, FL, USA.
✉e-mail: sumerlin@chem.ufl.edu

Published online: 24 November 2025

References

1. Friedrich, K. & Almajid, A. A. *Appl. Compos. Mater.* **20**, 107–128 (2013).
2. Xu, M. et al. *Compos. B Eng.* **251**, 110493 (2023).
3. Zhou, Y., Xu, Z. & Moore, J. S. *Nat. Synth.* https://doi.org/10.1038/s44160-025-00876-7 (2025).
4. Marquez, J. D. et al. *ACS Macro Lett.* **13**, 1345–1354 (2024).
5. Hughes, R. W. et al. *Angew. Chem. Int. Ed.* **63**, e202403026 (2024).
6. Siddiqi, Z. & Sarlah, D. *J. Am. Chem. Soc.* **143**, 21264–21269 (2021).
7. Fried, A. D., Wilson, B. J., Galan, N. J. & Brantley, J. N. *J. Am. Chem. Soc.* **144**, 8885–8891 (2022).
8. Fagnani, D. E., Kim, D., Camarero, S. I., Alfaro, J. F. & McNeil, A. J. *Nat. Chem.* **15**, 222–229 (2023).
9. Zhou, Y., Rodríguez-López, J. & Moore, J. S. *Nat. Commun.* **14**, 4847 (2023).
10. Gilchrist, G. C. et al. *J. Am. Chem. Soc.* **147**, 8398–8405 (2025).
11. Hsu, J. H., Ball, T. E., Oh, S., Stache, E. E. & Fors, B. P. *Angew. Chem. Int. Ed.* **63**, e202316578 (2024).

Competing interests

The authors declare no competing interests.