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Polymer recycling by mechanoradical capture

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Mechanoradicals are known to be sources of damage during polymer processing, and it remains challenging to stabilize, repair or utilize them. Recently, mechanoradicals formed during chain scission were intercepted directly by bis(butyl trithiocarbonate) in the solid state, redirecting the degradation pathways to repolymerization (repair) and complete depolymerization (recovery).

Mechanical recycling is a widely used method for processing plastic waste; however, in practice, heat and shear during processing lead to polymer degradation through backbone chain scission by the generation of highly reactive mechanoradicals. Besides efforts to stabilize or repair the broken chains caused by mechanoradicals, their intrinsic reactivity presents opportunities to redirect degradation pathways into useful transformations¹.

Now, a team led by Megan Hill at Colorado State University, USA, demonstrates that mechanoradicals generated by chain scission of polystyrene (PS) and poly(methyl methacrylate) (PMMA) during ball milling can be efficiently captured under solvent-free conditions by bis(butyl trithiocarbonate), yielding modified polymer fragments with trithiocarbonate (TTC) end groups (see Fig. 1)². The resulting TTC-functionalized polymer fragments can undergo chain extension to generate higher molecular weight polymers via light-mediated controlled polymerization (repair route). In addition, TTC-functionalized PMMA fragments can be further depolymerized to monomers by ball milling alone at 220 °C (recovery route).

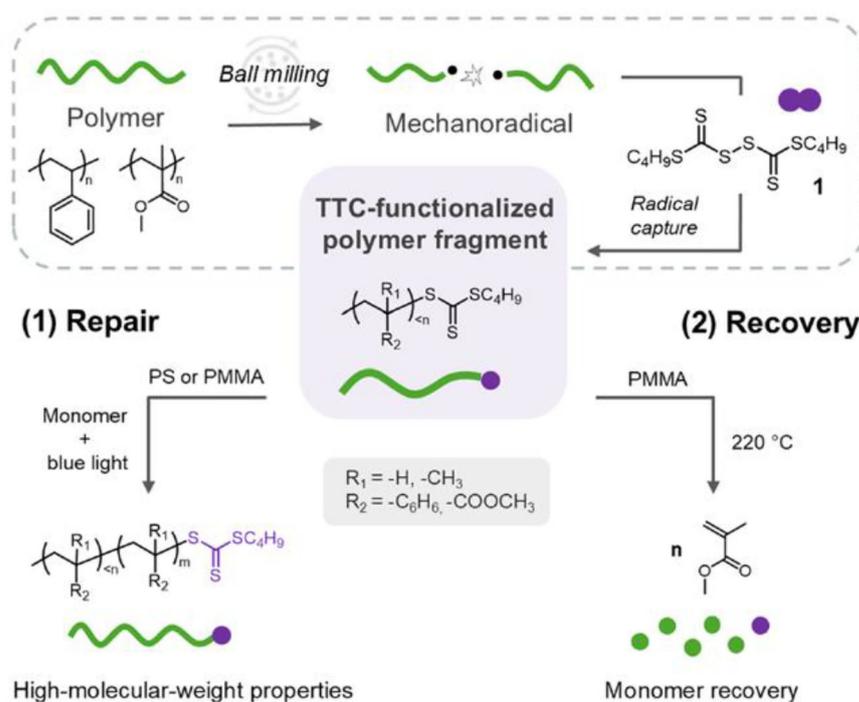
Employing photoiniferter polymerization, in which blue light directly activates the TTC end groups, TTC-functionalized PS and PMMA fragments undergo chain extension to achieve products with molecular weights equal to or exceeding those of the pristine polymers. In

addition, this “capture-and-repair” process can be repeated over three iterative cycles.

Inspired by recent studies demonstrating that thiocarbonylthio-end groups can facilitate lower depolymerization temperatures for PMMA^{3,4}, the team additionally finds that TTC-functionalized PMMA fragments produced via mechanoradical capture can similarly enable depolymerization to monomers at reduced temperatures, and that, surprisingly, ball milling alone can significantly enhance depolymerization even in the absence of bis(butyl trithiocarbonate).

“A major challenge was figuring out ball milling conditions and how to quantify the TTC functionalization,” comments Hill. “This was a new technique to our lab and it became clear that small variations in jar material, ball size, milling time, or even the texture of the polymer could drastically change how much the polymer degraded and therefore how many radicals we had to work with.” After a lot of experimentation, the team was able to make the conditions

Fig. 1 | Mechanoradical capture for polymer recycling. Mechanoradicals are generated by ball milling and captured by bis(butyl trithiocarbonate), yielding TTC-functionalized polymer chains, in turn enabling the “repair” and restoration of pristine polymer properties via chain extension (route 1) or the “recovery” of monomers via reduced temperature depolymerization (route 2). Reprinted with permission from *Journal of the American Chemical Society* <https://doi.org/10.1021/jacs.5c13585> (ref. 2). Copyright (2025) American Chemical Society.



reproducible and quantitatively correlate the degree of radical capture with downstream material properties.

“Our next step is to understand how broadly this strategy can be applied and if mechanoradical capture can introduce compatible chemical handles across different polymers. We are also working to understand how to harness these reactive intermediates in a more controlled way. Long term, we hope this chemistry will integrate with existing recycling technologies to provide a practical solution that improves outcomes rather than requiring entirely new infrastructure,” concludes Hill.

Huijuan Guo ✉

✉ e-mail: huijuan.guo@nature.com

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