

Environmentally Friendly Sustainable Thermoset Vitrimer-Containing Polyrotaxane for Circular Economy

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Durable green plastics with extended lifespans have self-healing properties and shape memory and are chemically recyclable and marine biodegradable. These plastics are attracting attention in applications such as adhesives and carbon fiber composite materials. Vitrimers satisfy many of these requirements, in terms of reprocessability and self-healing. Herein, we report significant improvements in the properties of vitrimers induced by the ester-exchange reaction between polyester-grafted polyrotaxane (PR) and the ester bonds of epoxy resin vitrimer to achieve the uniform molecular dispersion of PR. The optimal epoxy resin vitrimer incorporated with PR (VPR) exhibited enhanced toughness, with an elongation at break 5.3 times greater than that of a vitrimer devoid of PR; it also showed self-healing properties, was chemically recycled 10 times faster, and was recovered twice as fast. These results can be ascribed to the sliding diffusion motion of the PR, which lowers the energy required for transesterification. Furthermore, VPR showed 25 wt% biodegradation following exposure to seawater for 30 days. These findings may lead to the development of environmentally friendly plastics that exhibit most of the properties required for a circular economy.

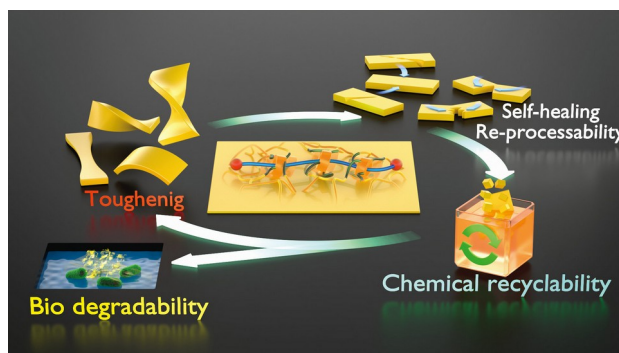


Figure. Vitrimer-containing polyrotaxane for circular economy.

Keywords: Polyrotaxane, Epoxy resin vitrimer, Dynamic covalent bond

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References

- [1] D. Montarnal, et al., Silica-like malleable materials from permanent organic networks. *Science* **2011**, 334, 6058, 965–968
- [2] G. Li, et al., Efficient mechanical toughening of polylactic acid without substantial decreases in stiffness and transparency by the reactive grafting of polyrotaxanes. *Journal of Inclusion Phenomena and Macrocyclic Chemistry* **2019**, 93:107–116
- [3] S. Pruksawan, et al., Homogeneously Dispersed Polyrotaxane in Epoxy Adhesive and Its Improvement in the Fracture Toughness. *Macromolecules* **2019**, 52, 6, 2464–2475.
- [4] S. Ando, et al., Environmentally Friendly Sustainable Thermoset Vitrimer-Containing Polyrotaxane. *ACS Mater. Lett.*, **2023**, 5, 12, 3156–3160.