

Supporting Information

**Achieving High Elasticity of Trans-1, 4-Polyisoprene with a
Combination of Radiation Crosslinking and Thiol-ene Grafting**

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Section 1. Preparation of irradiated TPI

Synthetic TPI pellets were provided by Qingdao Junxiang Technology Co. Ltd and without further purification. The Mooney viscosity is $59 M^{L^{100}_{3+4}}$ (100 stands for 100 °C, 3 + 4 stands for preheating for 3 min and shear for 4 min with a rate of 2 rpm). The trans-structure content in the sample is more than 95 %, as determined by $^1\text{H-NMR}$.

TPI films with 1 mm in thickness were prepared by compression molding at 120 °C under 10 MPa. The TPI plaques were cut into belts with a dimension of $12 \times 2.5 \text{ cm}^2$. The belts were sealed in glass tubes with a diameter of 3.0 cm and a length of 20 cm. To ensure N_2 atmosphere, the glass tube was repeatedly evacuated and filled with high purity N_2 .

The TPI was irradiated by γ -ray generated by a Co^{60} source in the Department of Applied Chemistry, Peking University. The irradiation doses were 100 kGy, 200 kGy, 300 kGy, 500 kGy, 700 kGy, and 1000 kGy. The dose rate was $\sim 100 \text{ Gy/min}$.¹

Section 2. Additional characterization data

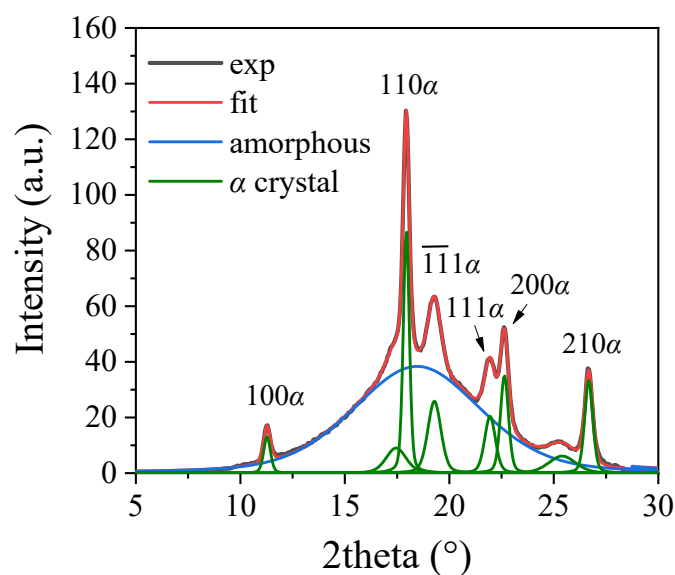


Fig. S1 Example illustrating the fitting of the WAXD profile of modified xTPI.

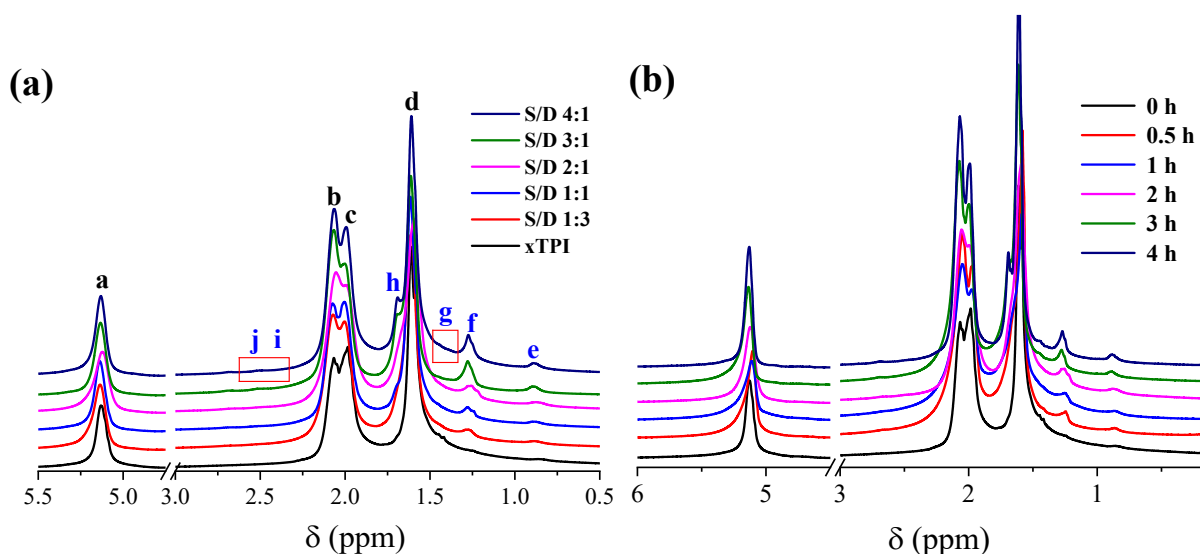


Fig. S2 (a) The ^1H -NMR spectra of modified xTPI vary with S/D ratios (fixed reaction time 2 h), (b) The ^1H -NMR spectra of modified xTPI vary with reaction times (fixed S/D 2:1).

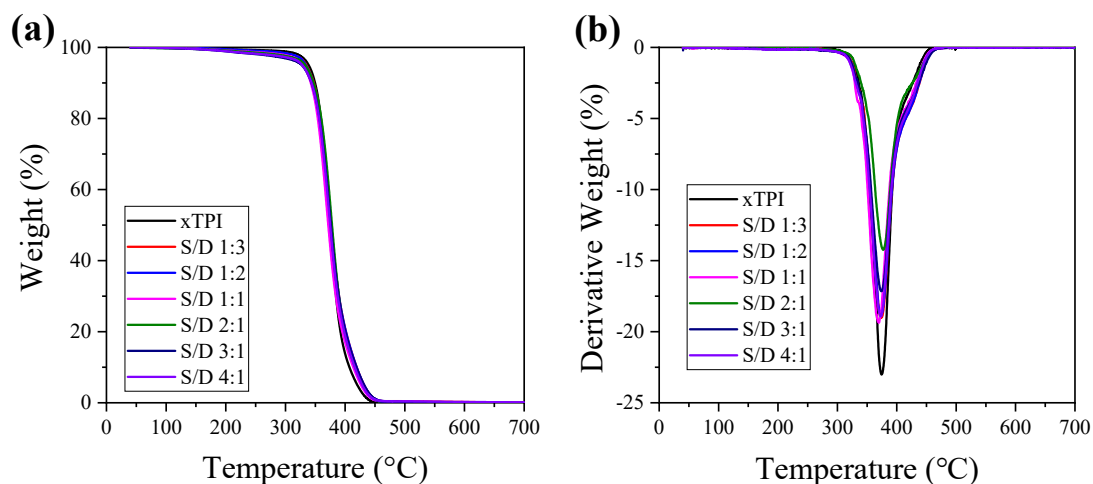


Fig. S3 (a) Thermal weight loss curves of g-xTPI with different S/D ratios (fixed reaction time 2 h). (b) The weight loss rate (DTG) curves of g-xTPI with different S/D ratios (fixed reaction time 2 h). The heating rate is $10\text{ }^\circ\text{C}/\text{min}$.

References

1. H. Zhao, C. Zhang, G. Liu, J. Li, B. Yang, H. Ma, X. Zhang and D. Wang, *Polymer Degradation and Stability*, 2022, **197**, 109869.