

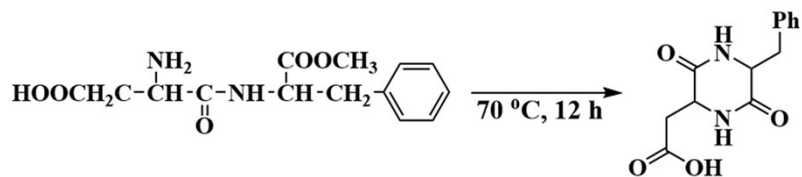
## Supporting Information

### **Molecularly Engineered Dual-Crosslinked Elastomer Vitrimers with Superior Strength, Improved Creep Resistance, and Retained Malleability**

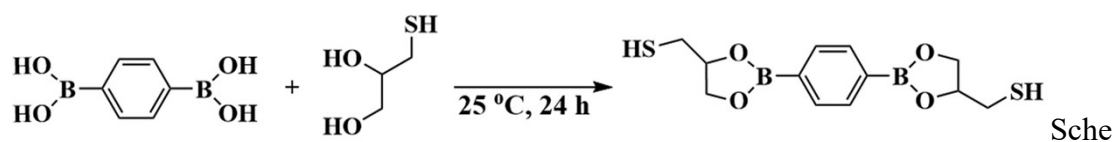
Lin Wang<sup>a</sup>, Yingjun Liu<sup>\*a, b</sup>, Yunhe Qiao<sup>a</sup>, Yuli Wang<sup>a</sup>, Ziwen Cui<sup>a</sup>, Shaoyi Zhu<sup>a</sup>,  
Fuwei Dong<sup>a</sup>, Sikun Fang<sup>a</sup>, Aihua Du<sup>\*a</sup>

<sup>a</sup>Key Laboratory of Rubber-Plastics (Ministry of Education), School of Polymer Science and Engineering, Qingdao University of Science and Technology, Qingdao 266042, China

<sup>b</sup>South China Advanced Institute for Soft Matter Science and Technology, South China University of Technology, Guangzhou 510641, China



Scheme S1. Synthesis of DKP.



Scheme S2. Synthesis of BDB.

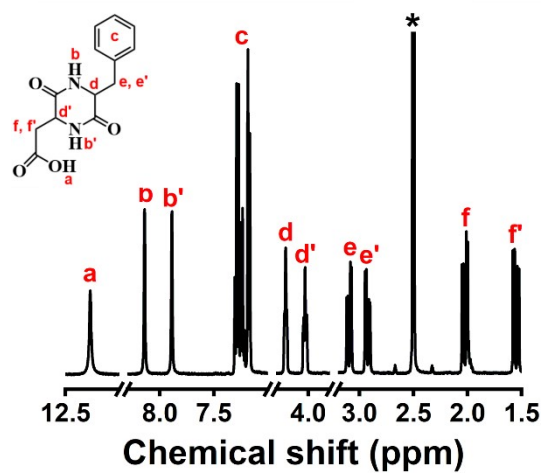


Figure S1. The  $^1\text{H}$  NMR spectrum of DKP. \*:  $\text{DMSO-}d_6$ .

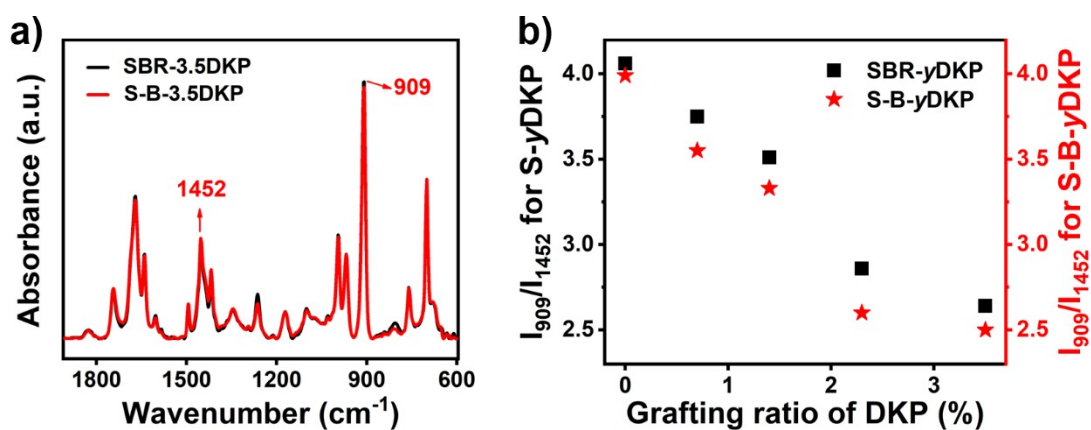


Figure S2. (a) The FTIR spectra of SBR-3.5DKP and S-B-3.5DKP; (b) The peak intensity ratio ( $I_{909}/I_{1452}$ ) for SBR- $\gamma$ DKP and S-B- $\gamma$ DKP.

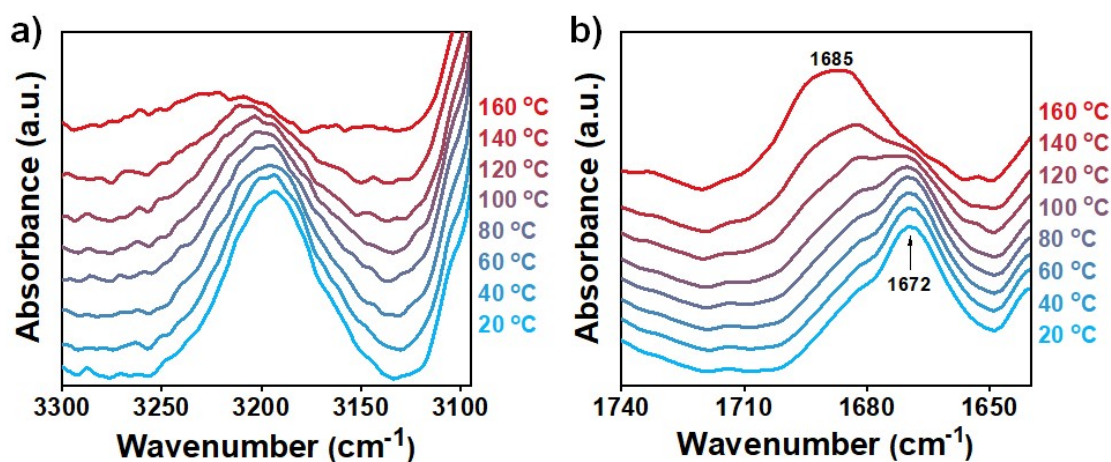


Figure S3. As in Figure 2 except the spectra were recorded as a function of decreasing temperature from 160 to 20 °C.

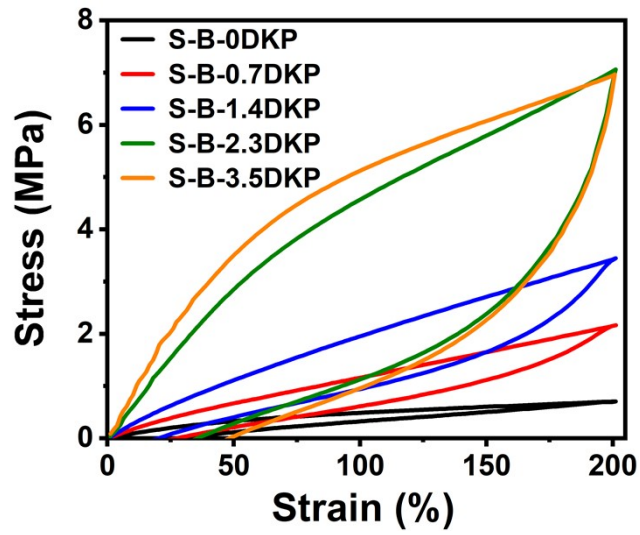


Figure S4. The first loading-unloading cycle of S-B-yDKP.

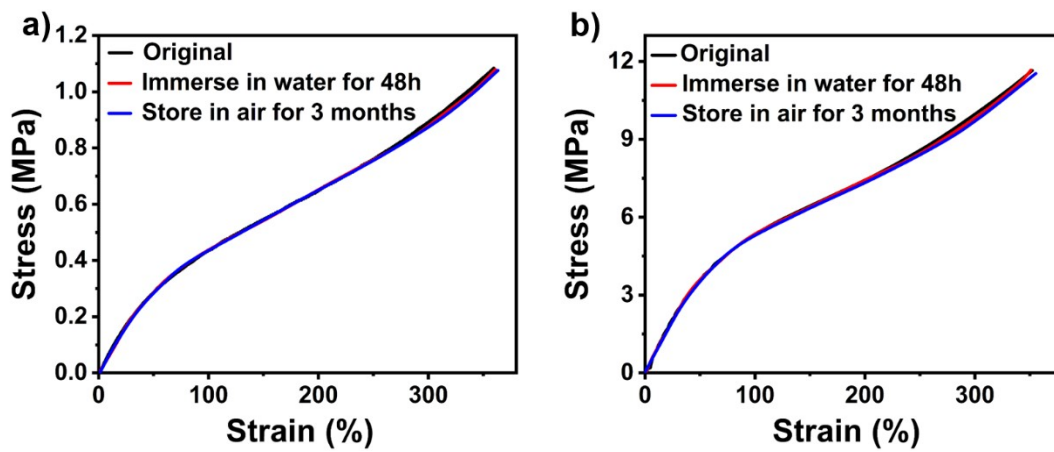


Figure S5. The stress-strain curves for original and water soaked (a) S-B-0DKP and (b) S-B-3.5DKP.

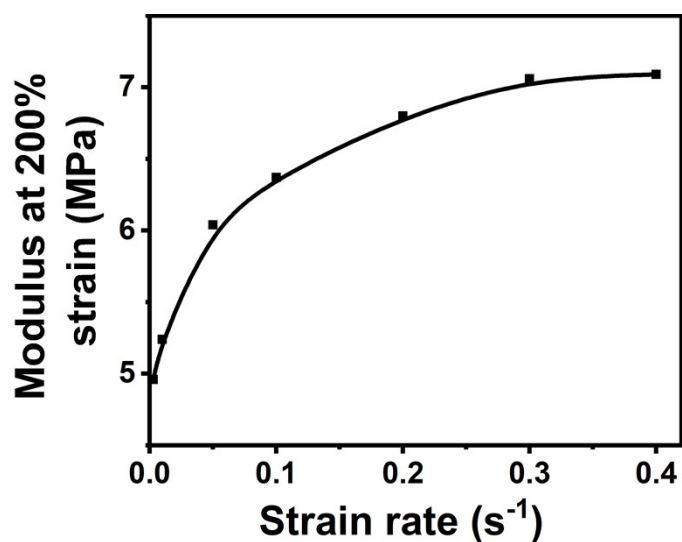


Figure S6. The modulus at 200% strain for S-B-3.5DKP at different strain rates.

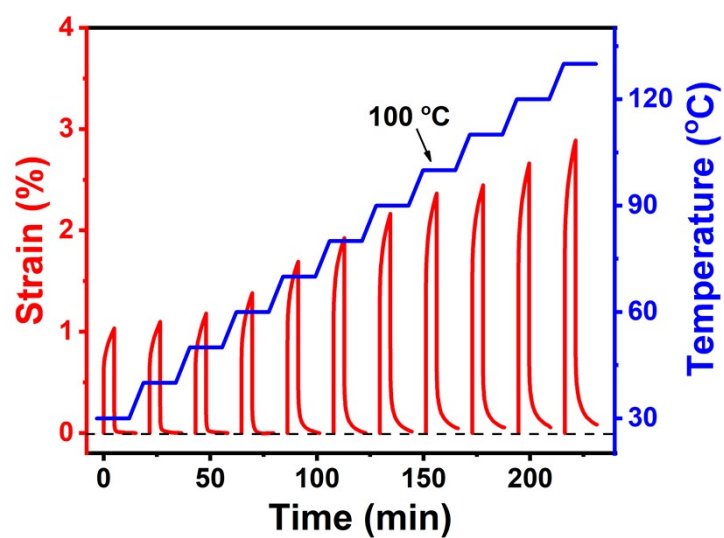


Figure S7. Cyclic strain/recovery profiles of S-B-1.4DKP during heating process with a stress of 0.03 MPa.

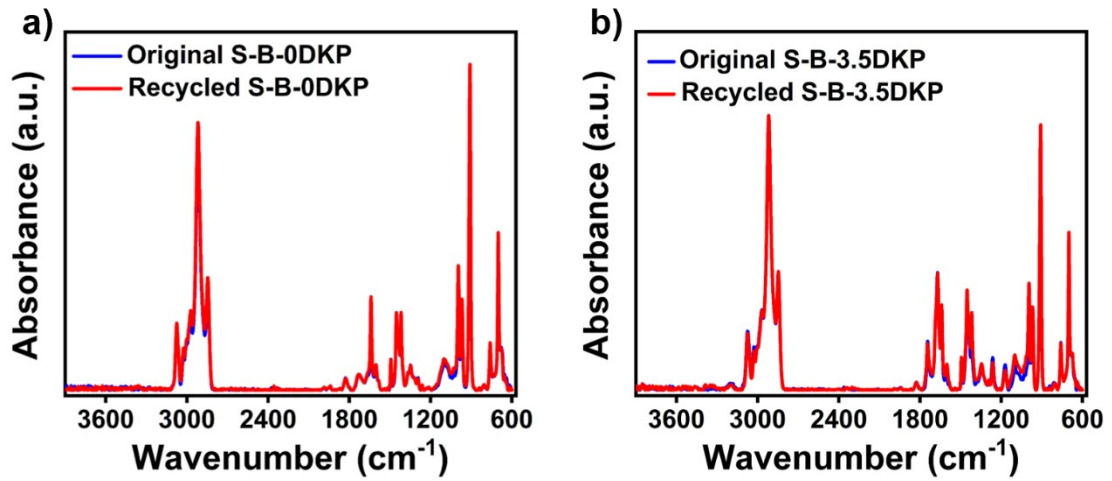


Figure S8. The FTIR spectra of original and recycled (a) S-B-0DKP and (b) S-B-3.5DKP.

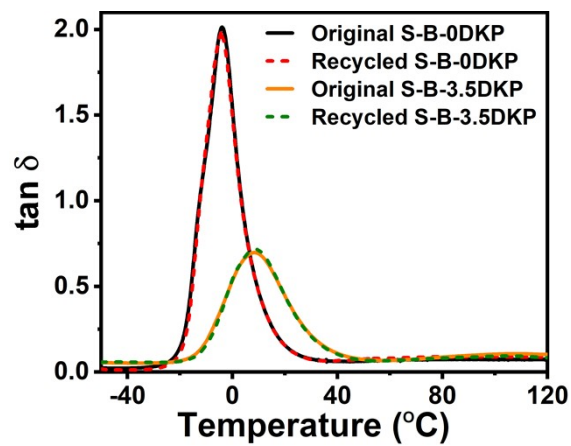


Figure S9. Temperature dependence of original and recycled (a) S-B-0DKP and (b) S-B-3.5DKP.

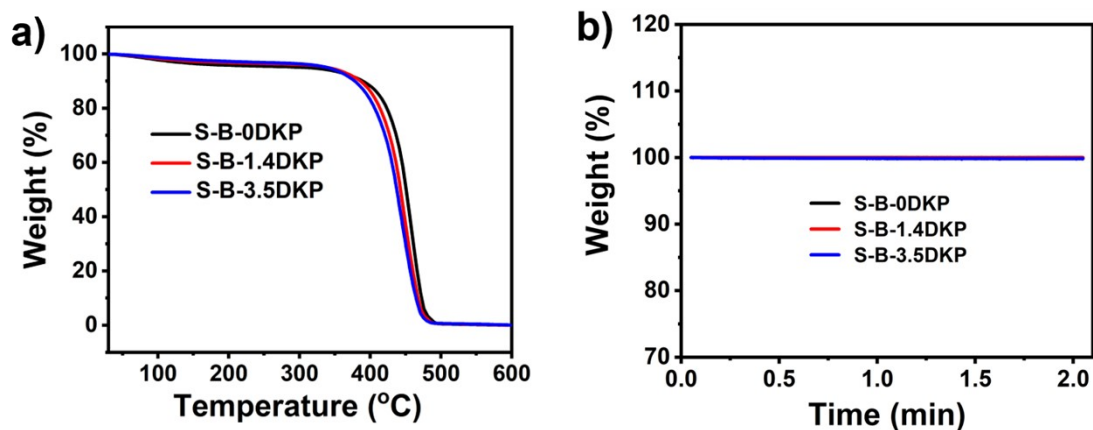


Figure S10. (a) TGA curves for S-B-0DKP, S-B-1.4DKP and S-B-3.5DKP; (b) Isothermal TGA curves of S-B-0DKP, S-B-1.4DKP and S-B-3.5DKP at 200 °C.

Table S1 Mechanical properties of S-B- $\gamma$ DKP.

Samples	Stress at 100% strain (MPa)	Ultimate stress (MPa)	Breaking strain (%)	Fracture energy (MJ/m <sup>3</sup> )
S-B-0DKP	0.47±0.06	1.10±0.26	352±55	2.05
S-B-0.7DKP	1.17±0.11	4.37±0.31	390±19	7.48
S-B-1.4DKP	1.97±0.06	5.90±0.61	328±49	11.54
S-B-2.3DKP	4.30±0.17	8.60±0.44	278±26	14.6
S-B-3.5DKP	5.07±0.38	10.2±1.87	308±24	22.79