Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information

Compatibility driven self-strengthening during radical-responsive remolding process of poly-isoprene vitrimers

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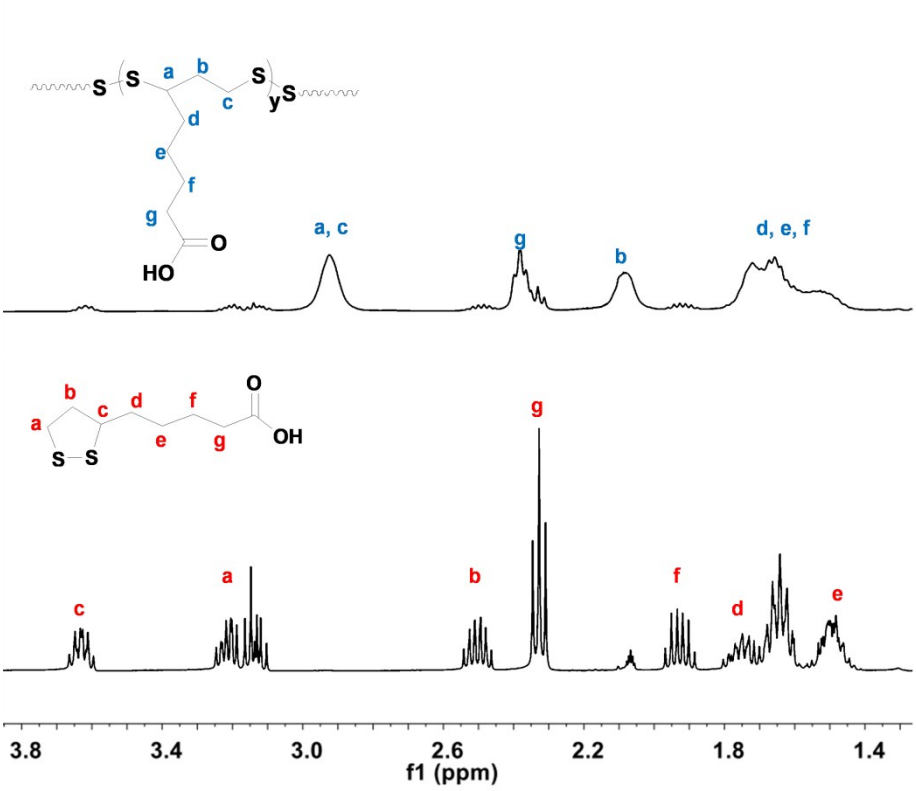

Figure S1. ^{1}H-NMR spectra of TA monomer and PTA in [D_6] actone.

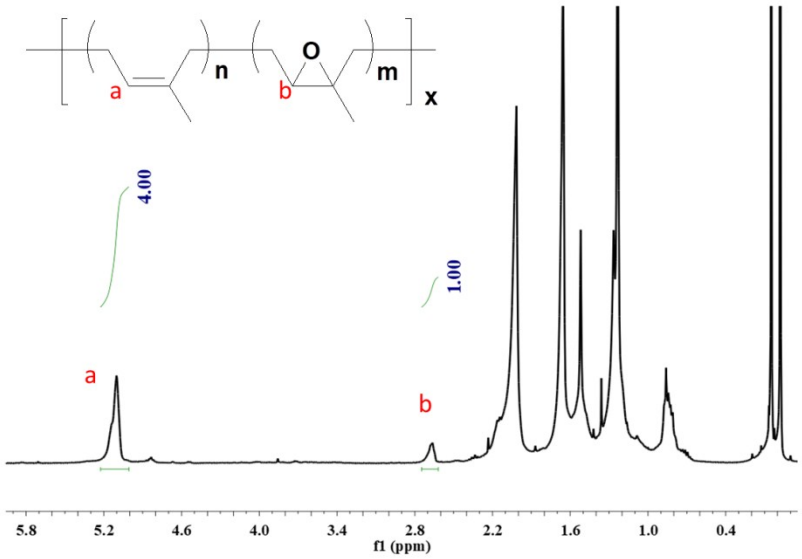

Figure S2. ^{1}H-NMR spectra of EPIP.

δ = 5.12 ppm for the olefinic hydrogen of cis-1,4 PIP; δ = 2.64 ppm for the epoxy hydrogen. The ratio of the segment with epoxy groups is 20% in the whole molecualr chain.

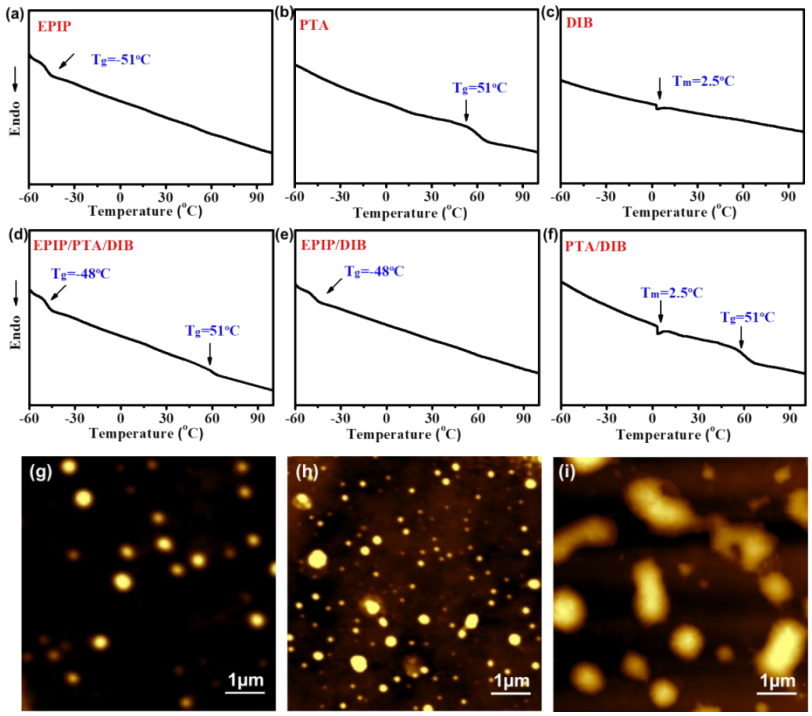

Figure S3. DSC charts of (a) EPIP, (b) PTA, (c) DIB, (d) EPIP/PTA/DIB mixture, (e) EPIP/DIB mixture, and (f) PTA/DIB mixture. AFM phase maps of (g) EPIP/PTA-4, (h) EPIP/PTA-8, and (i) EPIP/PTA-16.

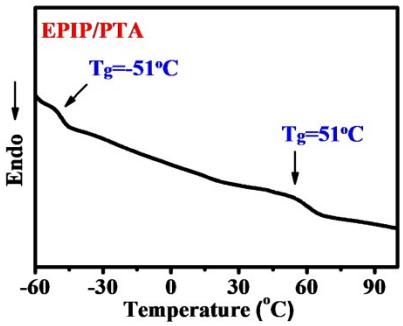

Figure S4. DSC chart of EPIP/PTA mixture.

Table S1. Formulations for EPIP/PTA-n and EPIP/SA samples

Samples	EPIP (g)	poly(carboxylic acid) (mg)	DIB (mg)	DMI (mg)	$Zn(Ac)_2$ (mg)
EPIP/PTA-4	10	309(PTA)	92	60	110
EPIP/PTA-8	10	619(PTA)	185	120	220
EPIP/PTA-16	10	1238(PTA)	370	240	440
EPIP/SA	10	517(SA)	0	120	220

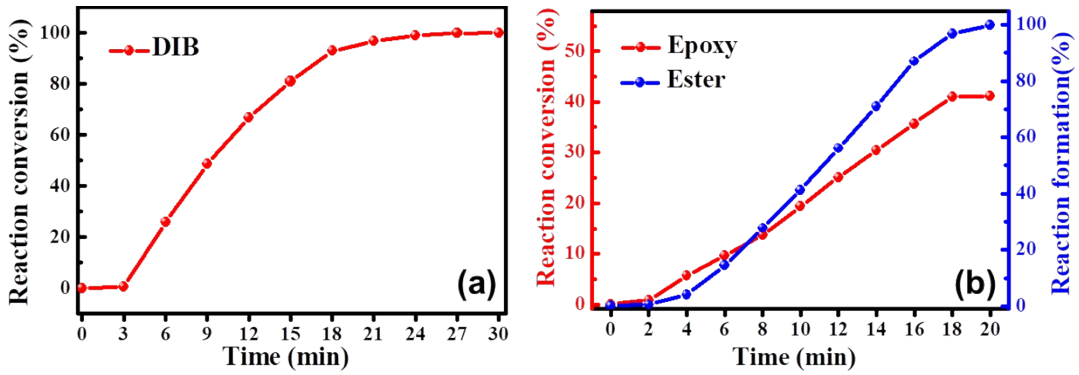

Figure S5. Reaction conversions calculated from the FTIR spectrum (Figure 1a and 1b). (a) Reaction conversion of DIB in PIP/PTA/DIB mixture. (b) Reaction conversion of oxirane rings and reaction formation of the β-hydroxyl ester bonds in EPIP/PTA-8.

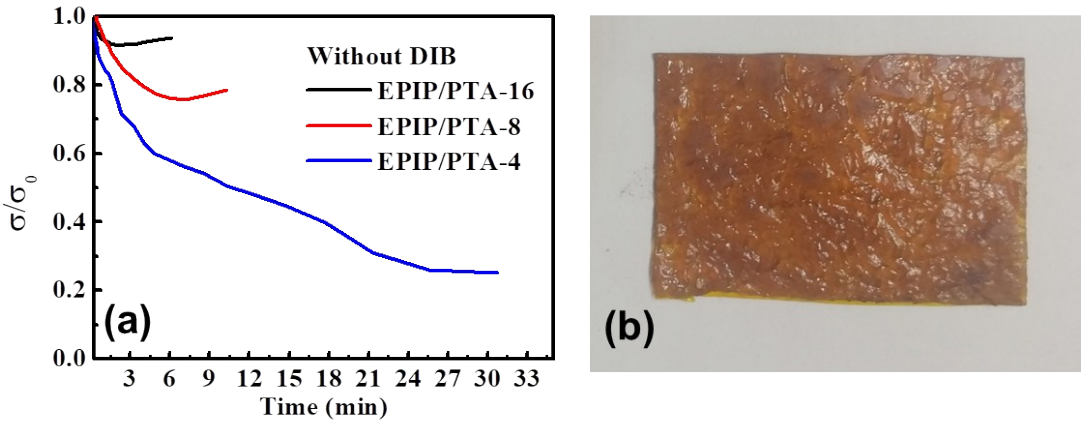

Figure S6. (a) Stress-relaxation curves of EPIP/PTA-n samples without DIB at 180 °C and the constant strain is 1%. (b) Photograph of the reprocessed rubber films without DIB by hot press at 180 °C.

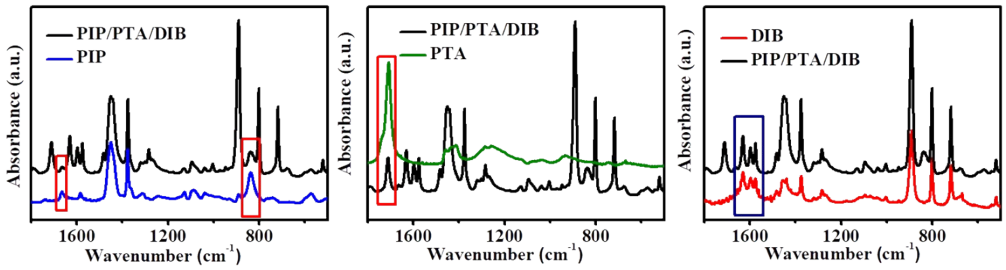

Figure S7. FTIR spectra of PIP, PTA, DIB and the PIP/PTA/DIB mixture at the initial state.

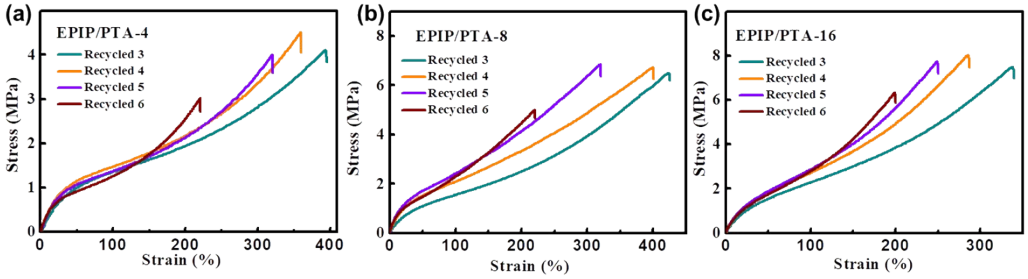

Figure S8. Stress-strain curves of the recycled samples of (c)EPIP/PTA-4, (d)EPIP/PTA-8 and (e) EPIP/PTA-16.

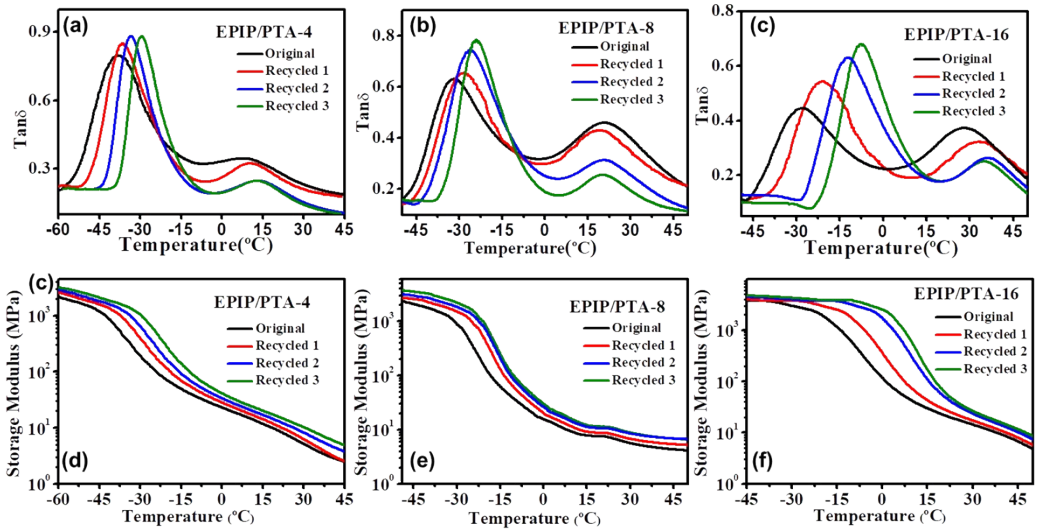

Figure S9. Temperature-dependent tan δ curves of the original and the recycled samples of (a)EPIP/PTA-4, (b)EPIP/PTA-8 and (c) EPIP/PTA-16. Storage modulus curves of the original and the recycled samples of (d)EPIP/PTA-4, (e)EPIP/PTA-8 and (f) EPIP/PTA-16.

Table S2. Crosslinking densities of the original and the recycled samples of EPIP/PTA-n

Crosslinking density 10^{-4}mol/cm^3	EPIP/PTA-4	EPIP/PTA-8	EPIP/PTA-16
Original	4	6	10
Recycled 1	6	9	15
Recycled 2	6.8	10	16.4
Recycled 3	7.2	10.8	17

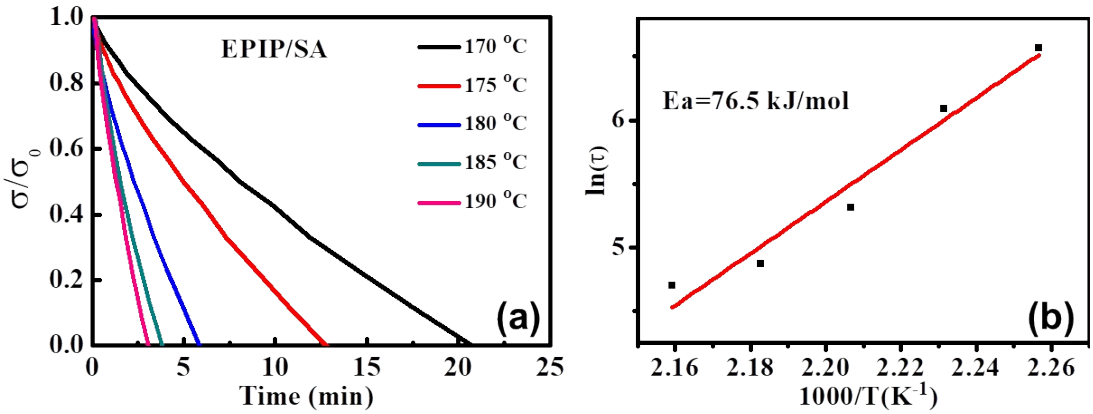

Figure S10. (a) Normalized stress relaxation for EPIP/SA at various temperatures ranging from 170 °C to 190 °C with a constant strain of 1%. (b) Fitting of τ* the Arrhenius equation for EPIP/SA.

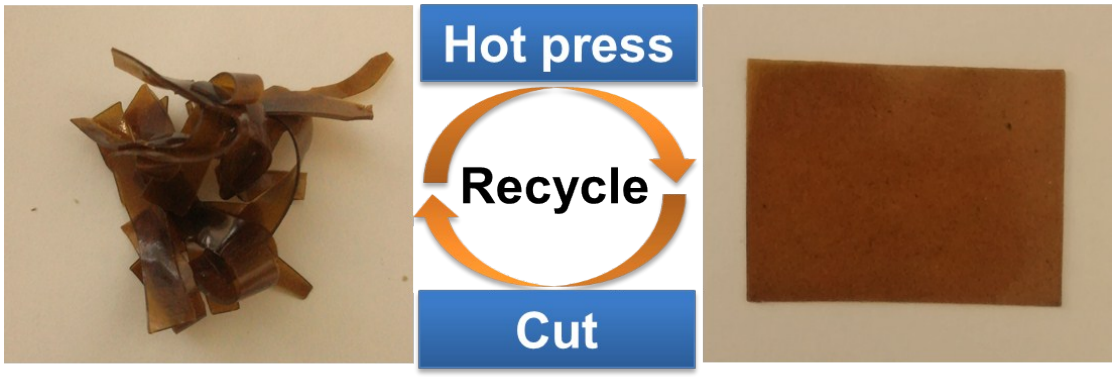

Figure S11. Photograph of the original cut samples (left) and the reprocessed (right) rubber films by hot press at 180 °C.

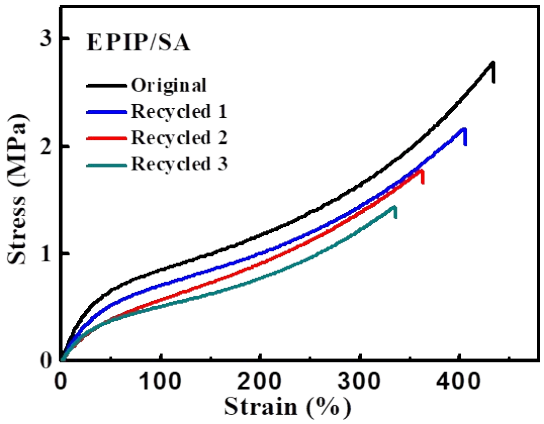

Figure S12. Stress-strain curves of the original and the recycled samples of EPIP/SA.