Supporting Information

Bioinspired Engineering of Sacrificial Metal-Ligand Bonds into Elastomers with Supra Mechanical Performance and Adaptive Recovery

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Table S1. Composition of the samples[a]

Codes	VPR (g)	sulfur (g)	zinc chloride/pyridine mole ratio	metal ions/pyridine mole ratio
VPR-*x*	100	1.5	variable (0, 0.25, 0.33, 0.5, 0.67)	
VPR-0.5-*y*	100	variable (0.2, 0.5, 1.0, 1.5, 2.5, 3.5)	0.5	
Ni(Co)-VPR	100	1.5		0.5
Fe(La)-VPR	100	1.5		0.33

[a] rubber ingredients: zinc oxide 5 g; stearic acid 2 g; dibenzothiazole disulfide 0.5 g; diphenyl guanidine 0.5 g; tetramethyl thiuram disulfide 0.2 g

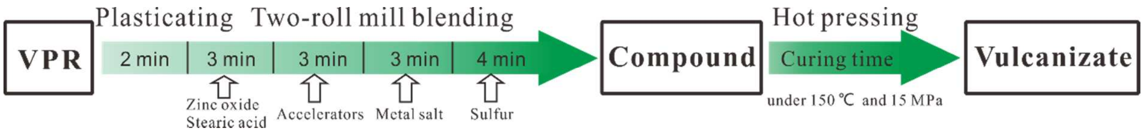

Figure S1. Flow chart for the preparation of VPR vulcanizates.

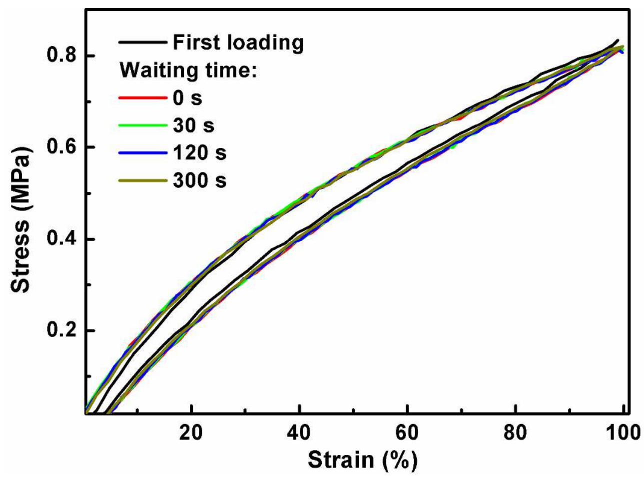

Figure S2. After the first cycle of loading and unloading, the recovery of VPR-0 after different waiting time at room temperature performed by cyclic tensile tests.

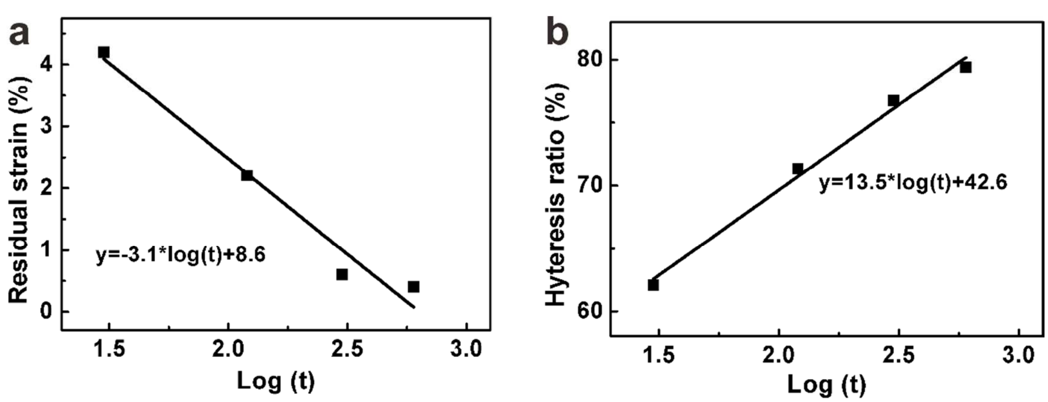

Figure S3. Plot for (a) residual strain and (b) hyteresis ratio versus the logarithm of *t*.

The straight line is a linearly fit to the data with the equation in the inset.

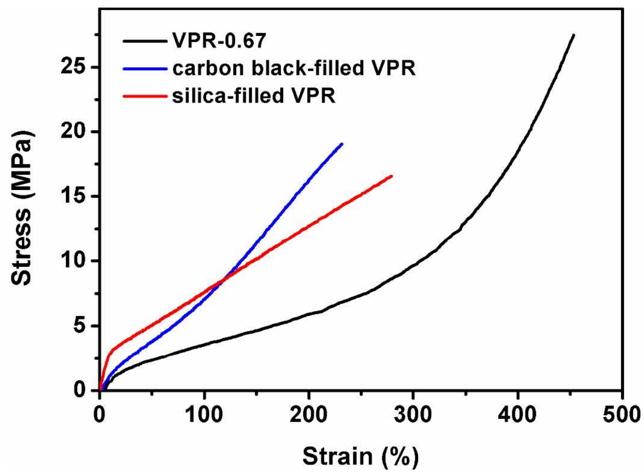

Figure S4. Comparison of the stress-strain curves for VPR-0.67, carbon black- and silica-filled VPR.

Table S2. Mechanical properties of VPR-*x*, CB- and silica-filled VPR

Samples	Young's modulus (MPa)	Stress at 100% strain (MPa)	Tensile strength (MPa)	Elongation at break (%)	Fracture energy (MJ/m^3)
VPR-0	4.9	0.8	3.5	428	6.3
VPR-0.25	6.2	1.3	8.9	526	17.4
VPR-0.33	12.7	2.0	13.3	495	23.4
VPR-0.5	15.4	2.9	20.4	453	30.6
VPR-0.67	19.8	3.7	27.8	454	38.7
carbon black-filled VPR	35.1	6.9	17.7	216	20.1
silica-filled VPR	38. 4	7.2	17.5	304	21.4

Table S3. Mechanical properties of VPR-0.5-*y* with different sulfur content. The mole ratio of $ZnCl_2$/VP is 0.5.

Samples	Young's modulus (MPa)	Stress at 100% strain (MPa)	Tensile strength (MPa)	Elongation at break (%)	Fracture energy (MJ/m^3)
VPR-0.5-0.2	7.6	1.9	24.2	615	44.3
VPR-0.5-0.5	9.3	2.1	25.6	579	37.3
VPR-0.5-1.0	13.1	2.9	22.6	474	35.5
VPR-0.5-1.5	15.4	2.9	20.4	453	31.5
VPR-0.5-2.5	23.9	5.1	18.8	323	28.8
VPR-0.5-3.5	27.8	6.2	18.7	273	23.6

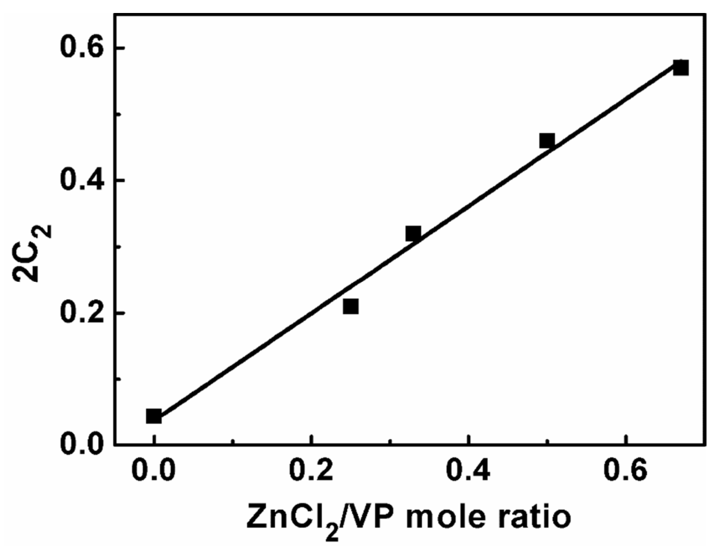

Figure S5. Plot for $2C_2$ versus $ZnCl_2$/VP mole ratio.

Figure S6. Appearance of the VPR-0.5-0 without sulfur.

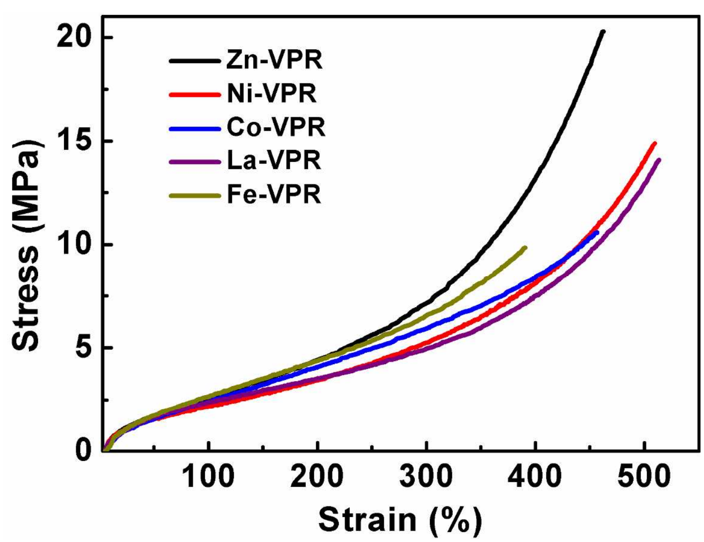

Figure S7. Typical stress-strain curves of VPR by incorporating different metal ions.

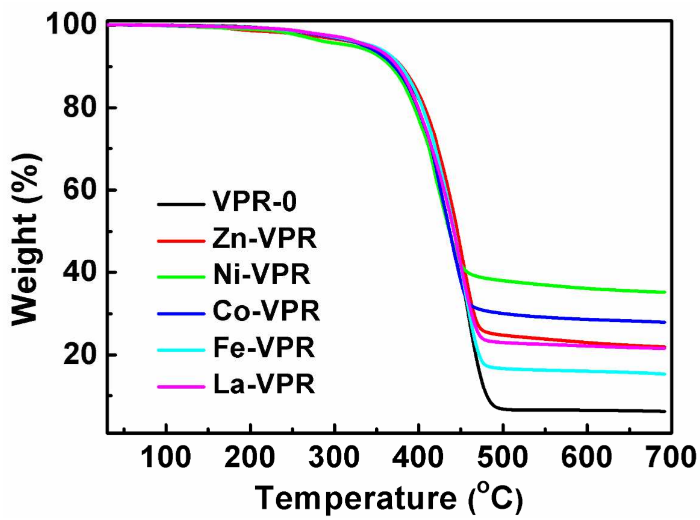

Figure S8. TGA curves of neat VPR and VPR samples with various metal ions.

Table S4. Mechanical properties of VPR using different metal ions.

Samples	Young's modulus (MPa)	Stress at 300% strain (MPa)	Tensile strength (MPa)	Elongation at break (%)	Fracture energy (MJ/m^3)
Zn-VPR	15.4	7.6	20.4	453	31.5
Ni-VPR	9.0	5.2	13.9	514	27.1
Co-VPR	11.4	5.6	11.6	486	22.1
La-VPR	6.7	4.9	14.1	513	26.3
Fe-VPR	10.5	6.6	9.9	391	17.8

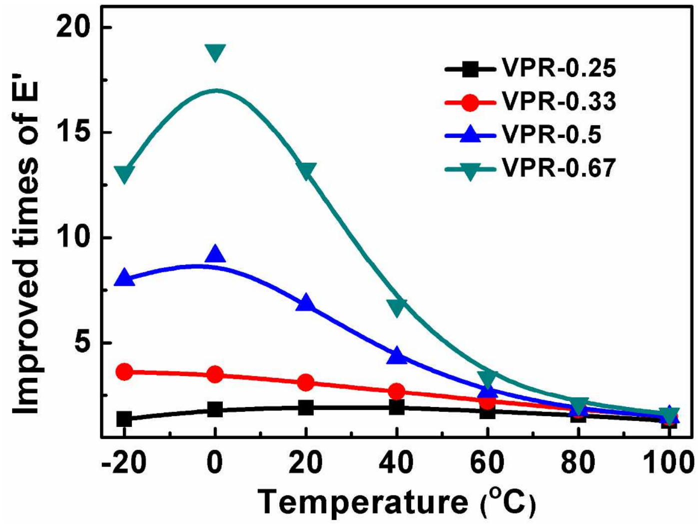

Figure S9. The E’ of VPR-*x* is normalized to that of VPR-0 at different temperature.