Supporting Information

Synergistic interfacial engineering of dynamic coordination and hydrogen bonding enables reprocessable ultra-strong rubber nanocomposites

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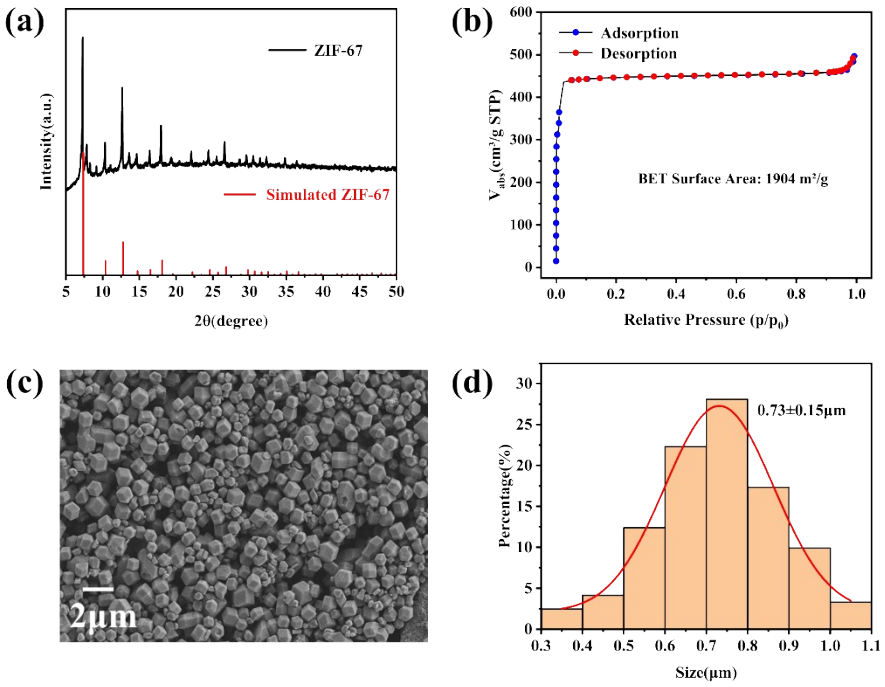

Figure S1. (a) XRD, (b) N_2 sorption curves, (c) SEM, (d) particle size distribution of ZIF-67

Table S1. Main formulations of HXNBRZ and HXNBRZ$_{10}$C.

Sample	Rubber/ g	ZIF-67/ g	CB/ g
HXNBRZ$_5$	20	1	0
HXNBRZ$_{10}$	20	2	0
HXNBRZ$_{15}$	20	3	0
HXNBRZ$_{10}$C$_{20}$	15	1.5	3
HXNBRZ$_{10}$C$_{30}$	15	1.5	4.5
HXNBRZ$_{10}$C$_{40}$	15	1.5	6

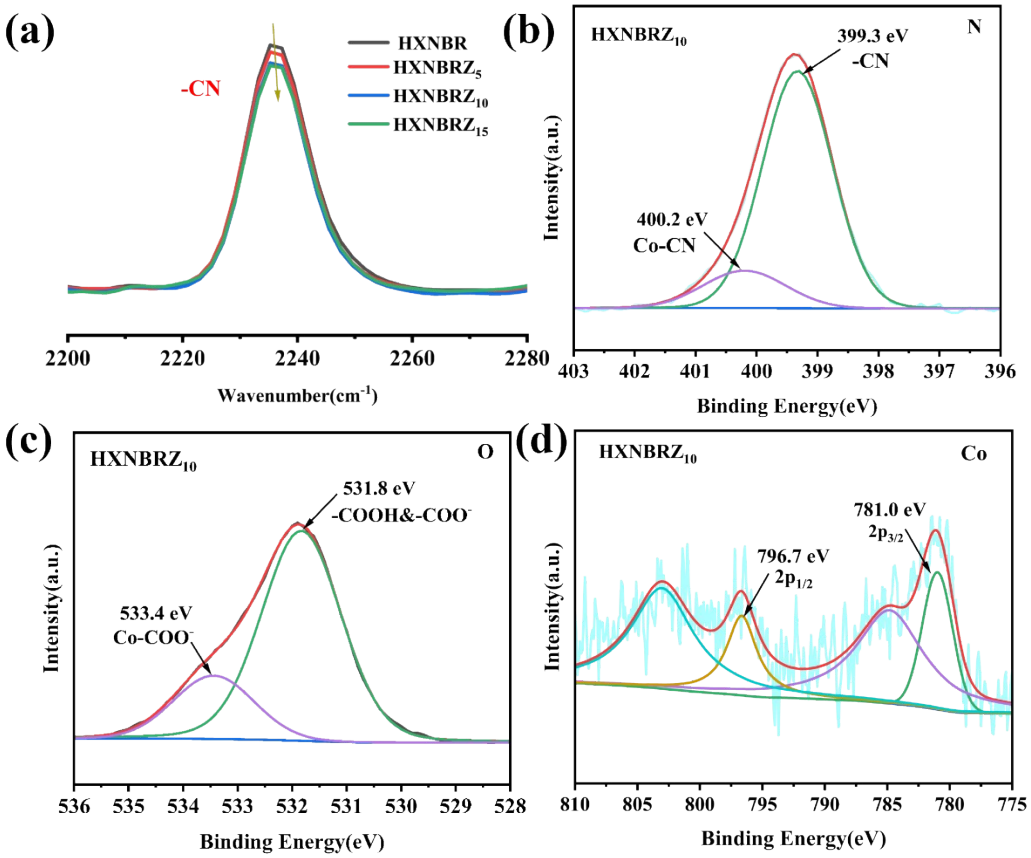

Figure S2. (a)FT-IR spectra of HXNBR and HXNBRZ. XPS spectra of HXNBRZ10: (b) N 1s, (c) O 1s, and (d) Co 2p.

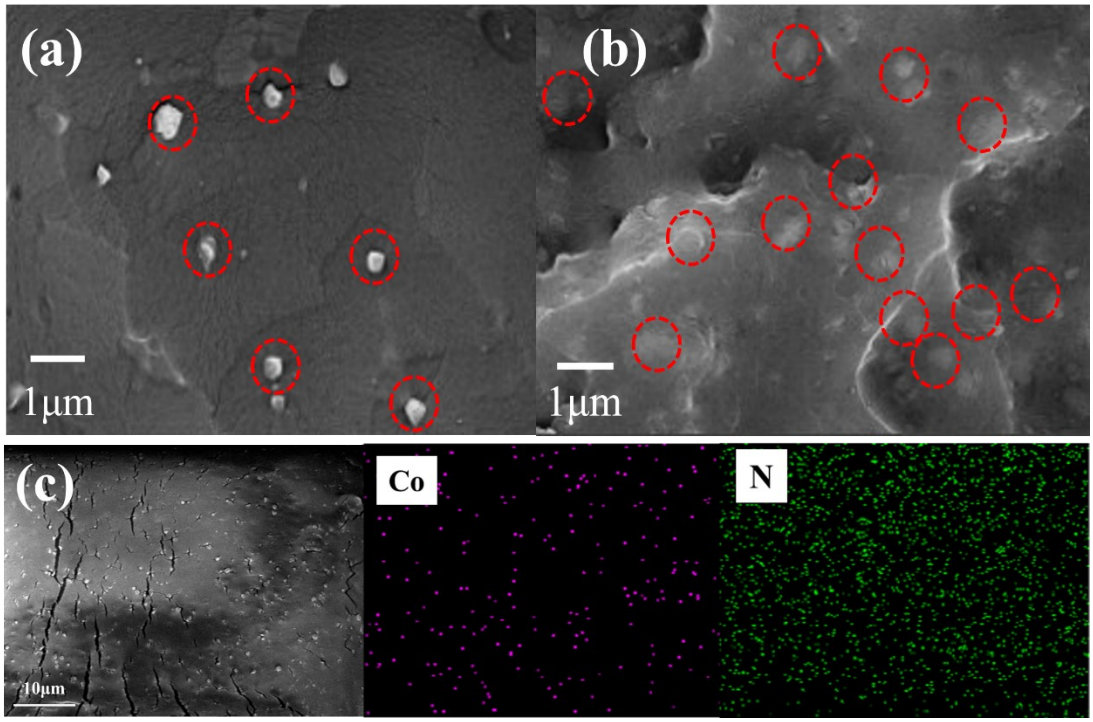

Figure S3. SEM of (a) $HXNBRZ_5$, (b)$HXNBRZ_{15}$. (c)EDS of $HXNBRZ_{10}$.

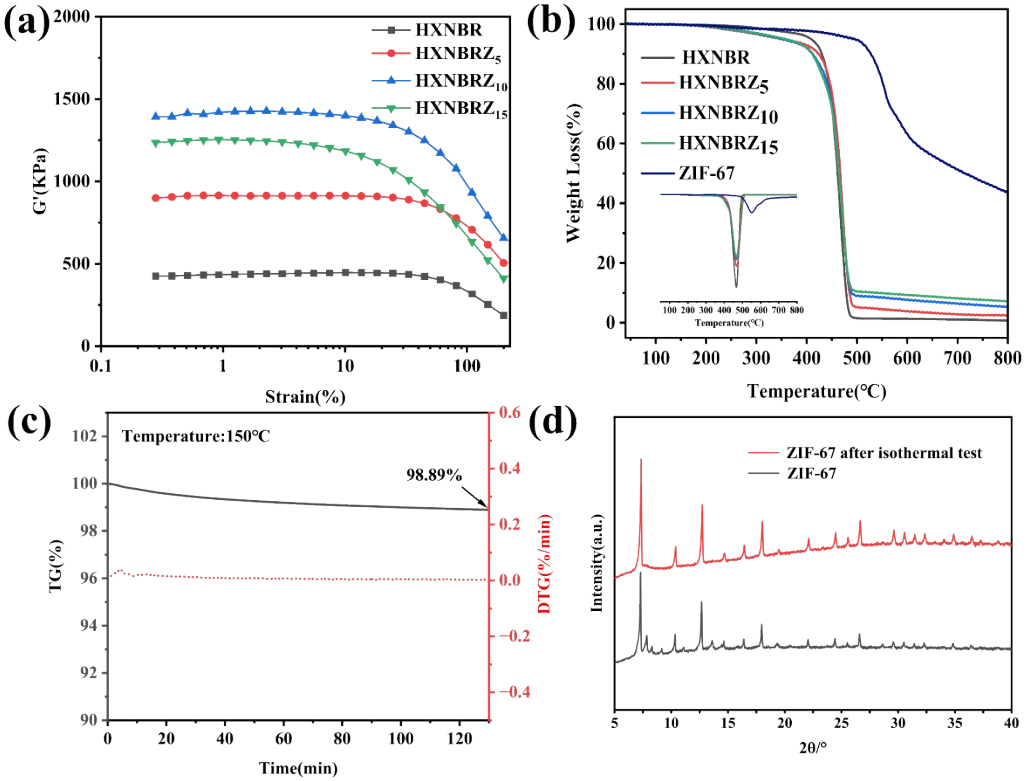

Figure S4. (a) RPA curves and (b) TGA curves of HXNBR and HXNBRZ. (c) isothermal TGA curve of ZIF-67. and (d) XRD pattern of ZIF-67 after the isothermal test.

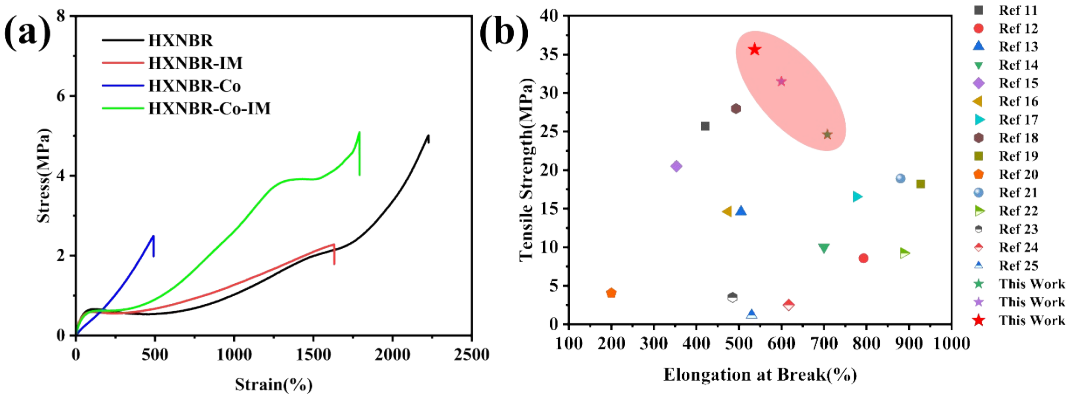

Figure S5. (a) Stress-strain curves of HXNBR, HXNBR-IM, HXNBR-Co and HXNBR -Co-IM (b) Comparison of tensile strength and elongation at break between this work and other reprocessable rubbers. [1-15]

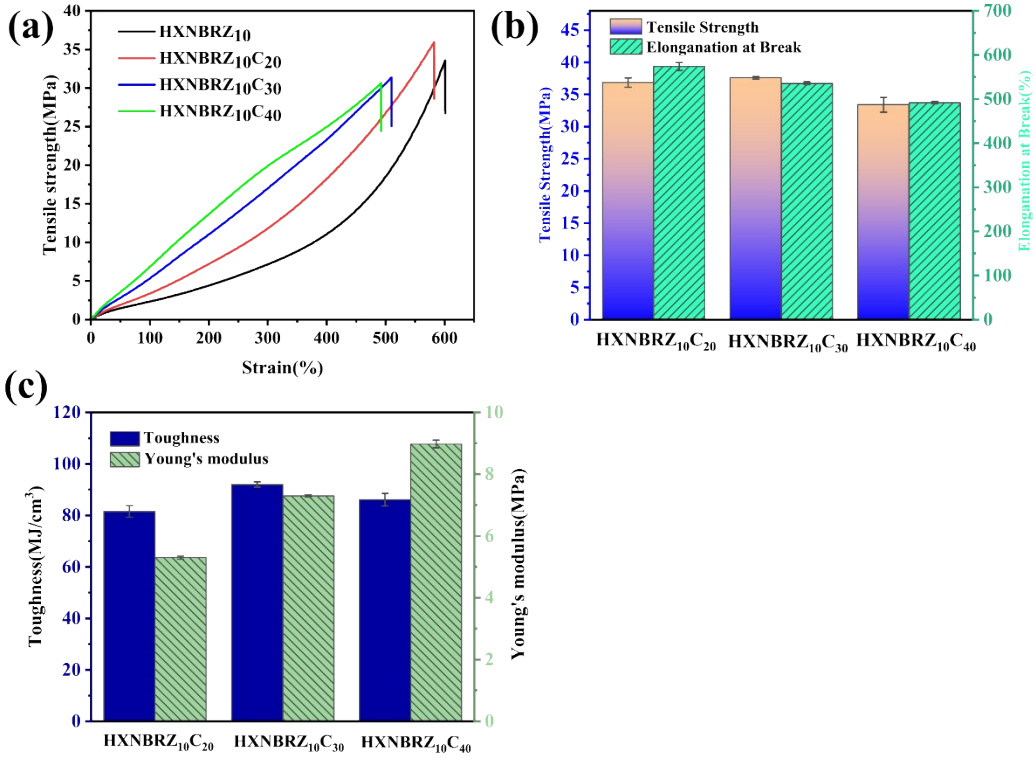

Figure S6. (a) Stress-strain curves, (b) Tensile strength and elongation at break, (c) Toughness and young's modulus of $HXNBRZ_{10}C_n$

Table S2. Mechanical properties of HXNBR, HXNBRZ and $HXNBRZ_{10}C$ films.

Sample	Tensile strength [MPa]	Strain [%]	Toughness [MJ m^{-3}]	Modulus [MPa]
HXNBR	5.0±0.01	2188±28	35.27±0.63	0.01±0.00
$HXNBRZ_5$	24.6±0.25	708±9	43.24±0.59	2.92±0.02
$HXNBRZ_{10}$	31.5±2.06	600±7	57.31±2.33	3.75±0.05
$HXNBRZ_{15}$	35.6±1.72	550±15	70.13±3.04	5.19±0.08
$HXNBRZ_{10}C_{20}$	37.8±2.56	590±12	81.50±2.24	5.30±0.05
$HXNBRZ_{10}C_{30}$	31.0±3.61	504±41	92.00±0.98	7.30±0.04
$HXNBRZ_{10}C_{40}$	31.1±1.52	496±4	86.11±2.43	8.98±0.12

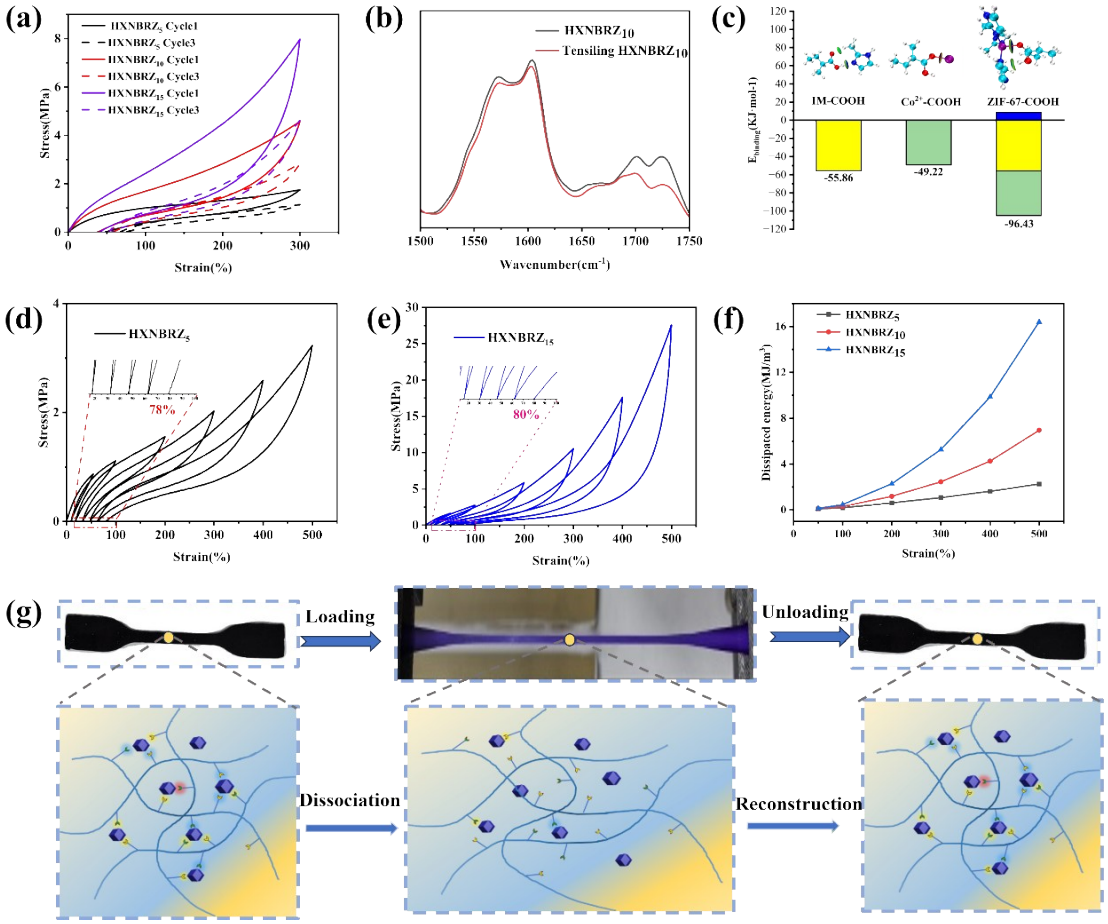

Figure S7. (a) 300% cyclic tensile curve of HXNBRZ. (b) FTIR spectra of static and tensiling $HXNBRZ_{10}$. (c) Binding energy of the -COOH with ZIF-67 synergistic interactions in HXNBRZ. Continuous cyclic tensile curve of (d) $HXNBR_5$, (e) $HXNBRZ_{15}$ with increasing strain without any stationary phase. (f) dissipated energy of HXNBRZ undergoing multiple consecutive increments. (g) Schematic diagram of the energy dissipation mechanism in HXNBRZ.

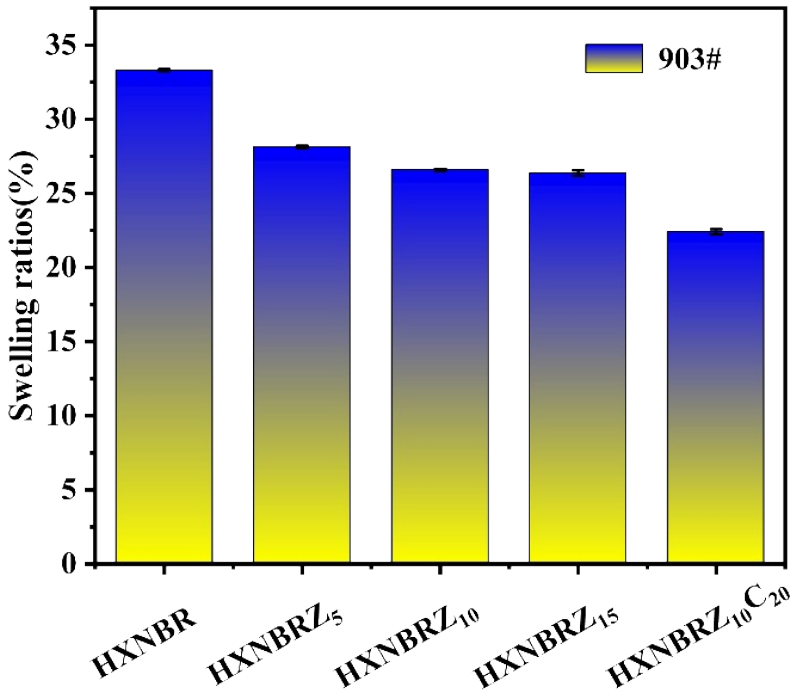

Figure S8. Oil resistance properties of HXNBR-DCP and HXNBRZ.

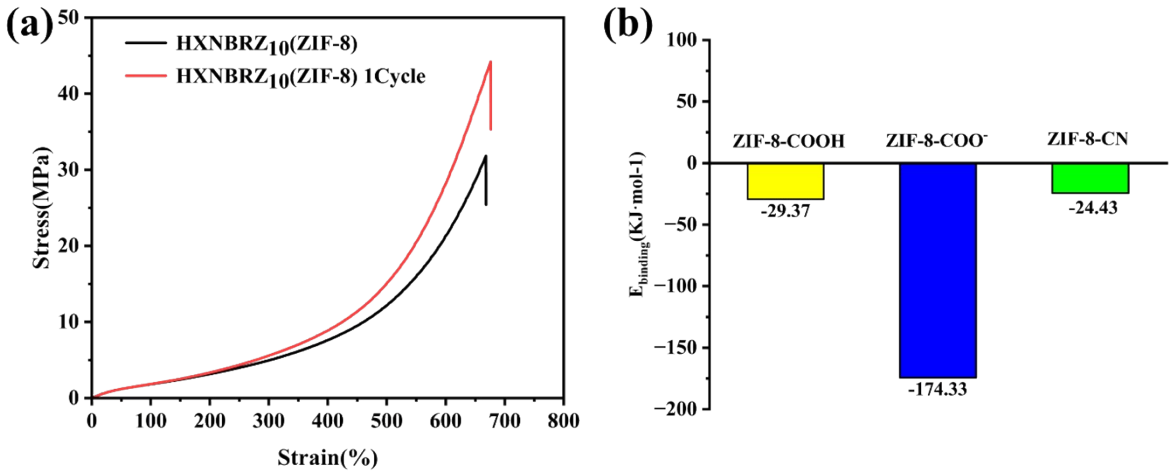

Figure S9 (a)Stress-strain curves of original and reprocessing HXNBRZ$_{10}$(ZIF-8). (b) Binding energy of groups with ZIF-67 synergistic interactions in HXNBRZ$_{10}$(ZIF-8).

Table S3. Mechanical properties of HXNBRZ 1Cycle.

Sample	Tensile stress [MPa]	Strain [%]	Toughness [MJ m^{-3}]	Modulus [MPa]
HXNBRZ$_5$ 1C	46.64±1.62	656±6	66.96±2.83	3.20±0.07
HXNBRZ$_{10}$ 1C	51.47±1.09	610±1	91.86±1.90	4.45±0.07
HXNBRZ$_{15}$ 1C	46.36±1.75	544±7	93.40±6.91	6.11±0.09
HXNBRZ$_{10}$C$_{20}$ 1C	42.62±1.58	521±14	95.02±6.06	6.99±0.10
HXNBRZ$_{10}$C$_{30}$ 1C	37.60±0.21	536±3	92.00±0.98	7.30±0.04
HXNBRZ$_{10}$C$_{40}$ 1C	33.41±1.17	492±3	86.11±2.43	8.98±0.12

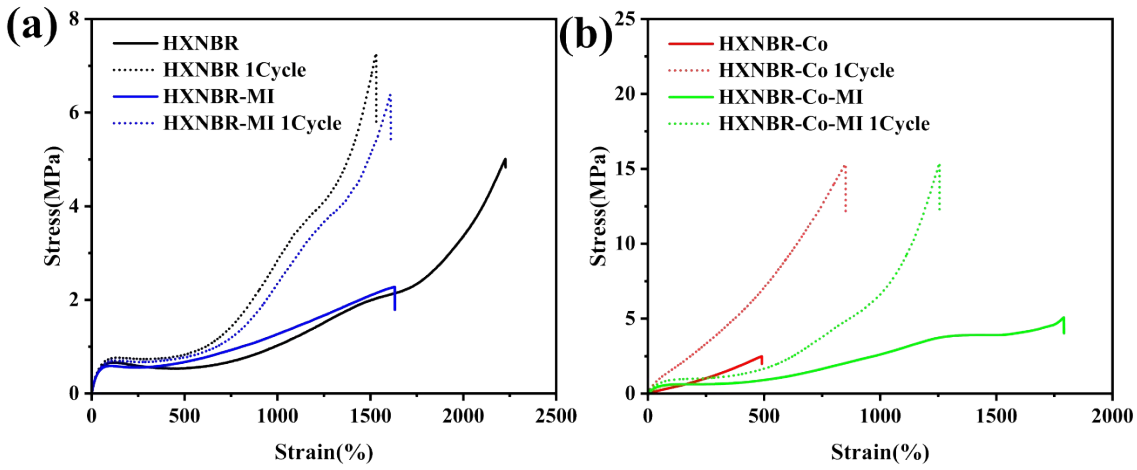

Figure S10 Stress-strain curves of original and reprocessing (a)HXNBR and HXNBR-MI, (b)HXNBR-Co and HXNBR-Co-MI.

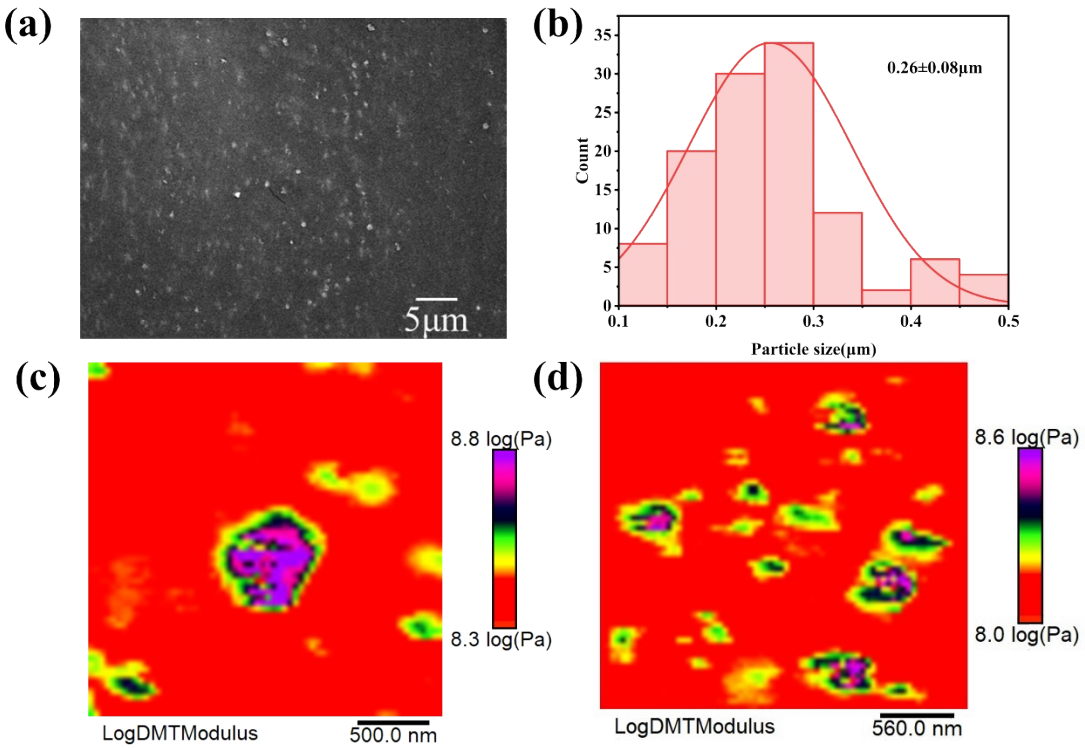

Figure S11. SEM of (a) $HXNBRZ_{10}$ 1Cycle, (b) particle size distribution of ZIF-67, AFM (c) $HXNBRZ_{10}$, (d) $HXNBRZ_{10}$ 1Cycle.

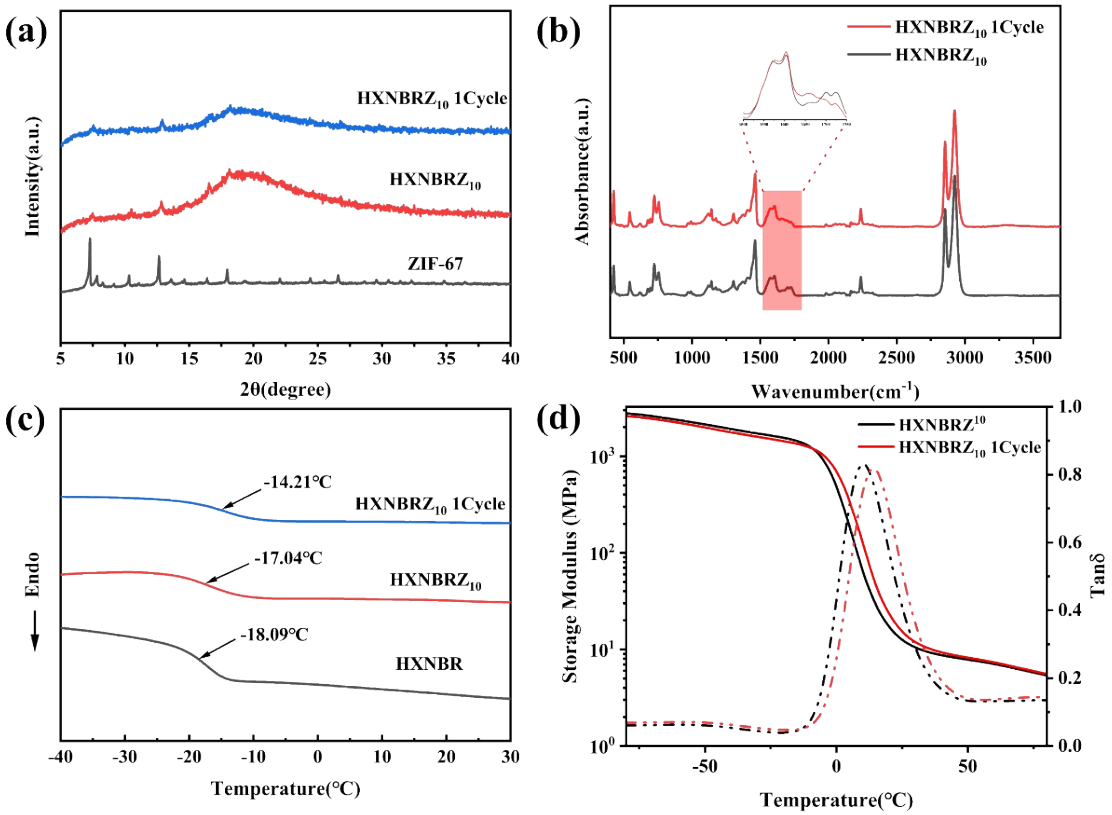

Figure S12. (a) XRD, (b) FTIR, (c) DSC, (d) DMA of $HXNBRZ_{10}$ and $HXNBRZ_{10}$ 1Cycle

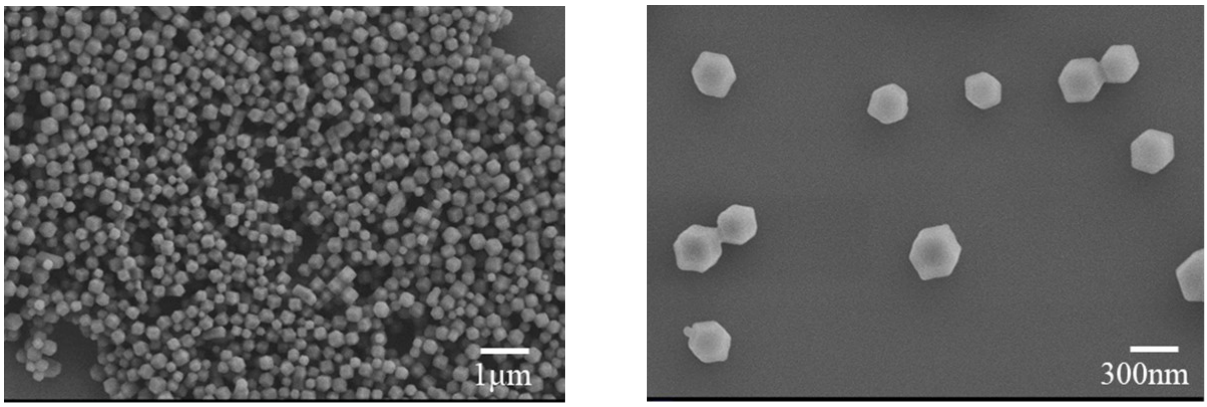

Figure S13. SEM of ZIF-67 with a particle size of 300nm

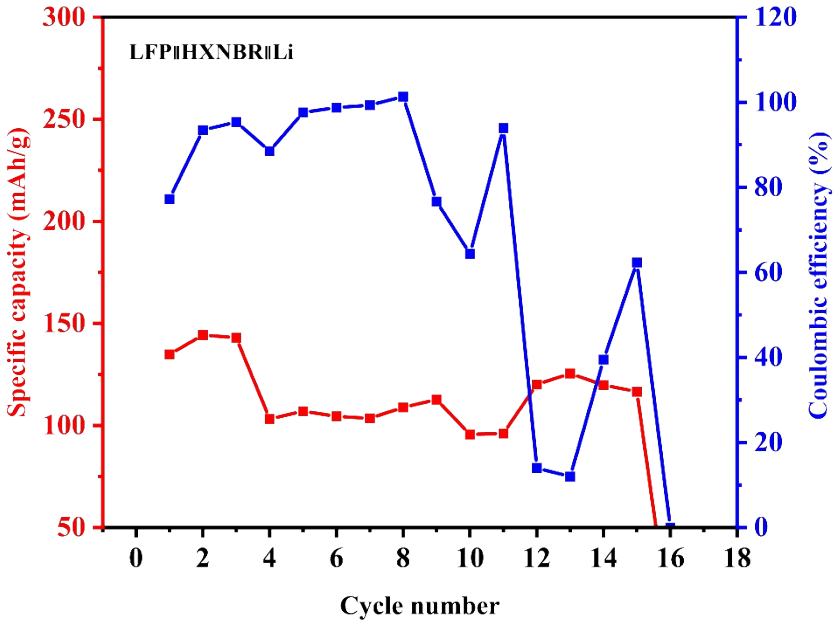

Figure S14 Cycling performance of the LiFePO4‖HXNBR‖Li cell at 0.2C at room temperature.

Table S4. Comparison of mechanical properties of rubbers after reprocessing between this work and other reprocessable elastomers.

Sample	Tensile stress [MPa]	Cycle	Recover ratio [%]	Ref
SBR	~15	1	~83	1
NBR	6.5	2	97	2
EEUG	~9	3	~92	3
HENR	~15	2	~92	5
ENR	4.8	3	76	6
ENR	9.3	2	93	7
ENR	10.5	2	81	8
ENR	11.84	2	65	9
PU	~28	2	~90	16
PU	~20	3	~89	17
PU	39.3	3	91	18
PU	~40	3	~93	19
PU	15.2	5	81	20
HXNBRZ$_{10}$C$_{20}$	45.2	5	128	This Work
HXNBRZ$_{10}$ (300 nm)	47.7	5	95	This Work

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