Supporting Information

Self-Healing Flexible Urea-g-MWCNTs/Poly(urethane-sulfide) Nanocomposite for Sealing Electronic Devices

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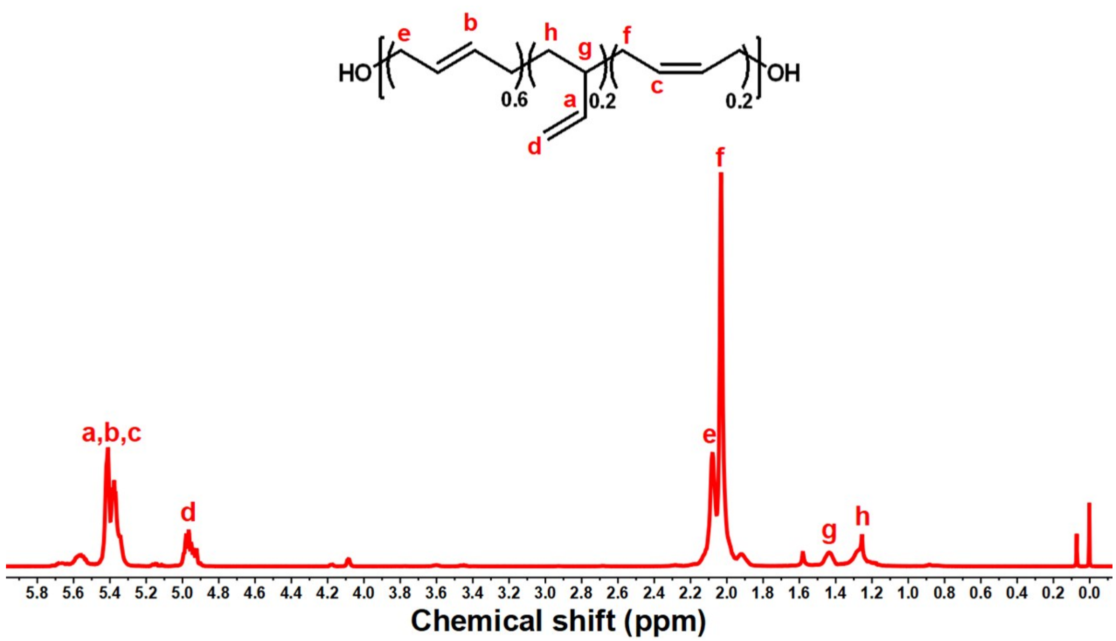

Figure S1. ^{1}H NMR spectrum of HTPB in $CDCl_3$ (400MHz).

In the ^{1}H NMR spectrum of HTPB, the peaks at 5.35-5.45 ppm correspond to the -CH= proton of the 1,2-vinyl units and 1,4-(cis and trans) units. The peak at 4.97 ppm is attributed to the $=CH_2$ proton of the 1,2-vinyl units. The peaks at 1.97-2.25 ppm are assigned to the $-CH_2-$ protons of the 1,4-(cis and trans) units. The peaks at 1.45 ppm and 1.26 ppm belong to the CH and $-CH_2-$ protons of 1,2-vinyl units[1-3].

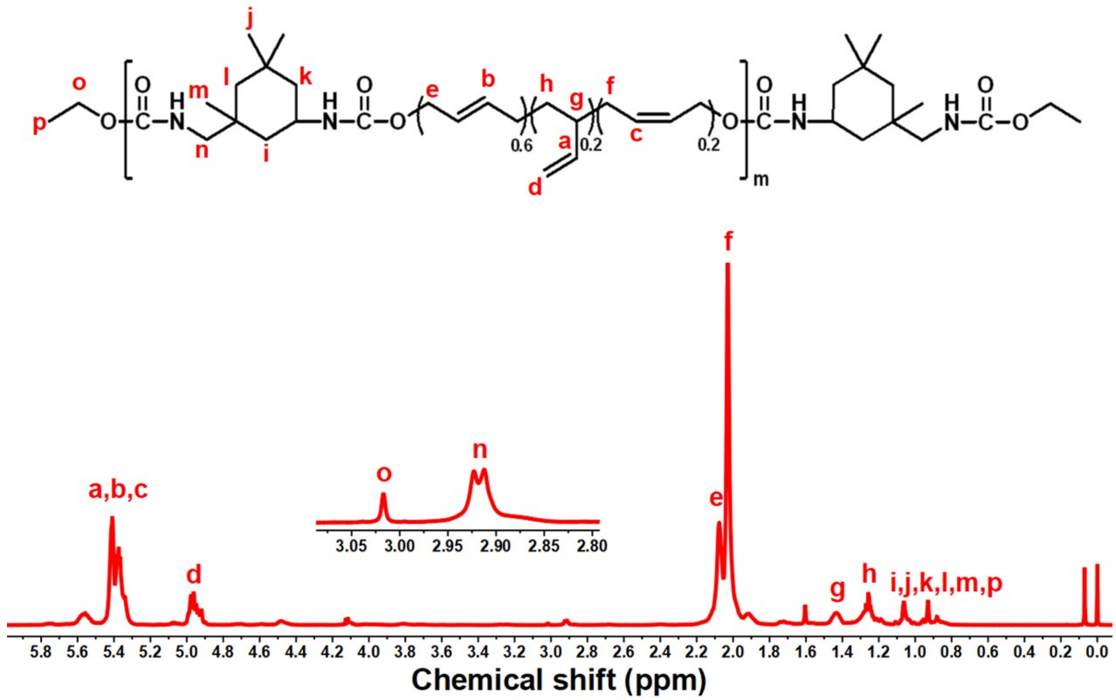

Figure S2. ^{1}H NMR spectrum of the ethanol-terminated polyurethane in $CDCl_3$ (400MHz).

Compared to the ^{1}H NMR spectrum of HTPB, the new peaks at 0.88-1.06 ppm for $-CH_3$ and $-CH_2-$ protons of IPDI were observed. The peaks at 2.90-2.94 ppm and 3.02 ppm are attributed to the methylene proton of $-CH_2-NH-COO-$ and methylene proton of ethanol, respectively[4-5].

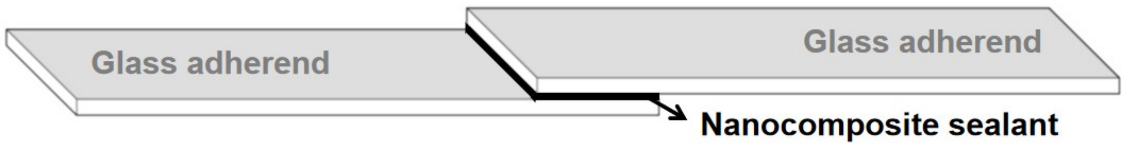

Figure S3. The model of lap-shear test for nanocomposite.

Table S1. Compositions and mechanical properties of nanocomposite

Samples	Compositions			Mechanical properties	
	Urea-g-MWCNTs (g)	Ethanol-terminated polyurethane (g)	*Crosslinker (g)	Tensile strength (MPa)	Breaking strain (%)
ETPU*	0	50	0	1.81±0.12	521±15
PUS2*	0	50	2	2.30±0.09	463±11
PUS4*	0	50	4	2.71±0.10	351±14
PUS6*	0	50	6	3.15±0.05	270±10
PUS8*	0	50	8	3.87±0.03	242±8
PUS10*	0	50	10	4.03±0.10	145±14
ETPU	0.15	50	0	2.12±0.09	515±14
PUS2	0.15	50	2	2.62±0.08	451±13
PUS4	0.15	50	4	3.01±0.13	344±17
PUS6	0.15	50	6	3.58±0.06	258±9
PUS8	0.15	50	8	4.03±0.10	234±13
PUS10	0.15	50	10	4.22±0.11	133±15
PUOD4	0.15	50	4	3.18±0.10	331±4
PUOD6	0.15	50	6	3.65±0.08	243±11
PUOD8	0.15	50	8	4.12±0.09	222±13

* PUOD4, PUOD6 and PUOD6 use 3,6-dioxa-1,8-octanedithiol as crosslinker, the other samples use polysulfide oligomer as crosslinker.

Table S2. TGA results of PUSχ*

Samples	$T_{1.5\%}$ (°C)	$T_{5\%}$ (°C)	$T_{50\%}$ (°C)	$T_{90\%}$ (°C)
PUS2*	211.3	229.2	401.8	445.4
PUS4*	214.5	237.9	404.5	447.6
PUS6*	218.8	250.3	409.1	455.4
PUS8*	221.9	259.0	413.3	458.2
PUS10*	225.2	267.7	415.4	460.7

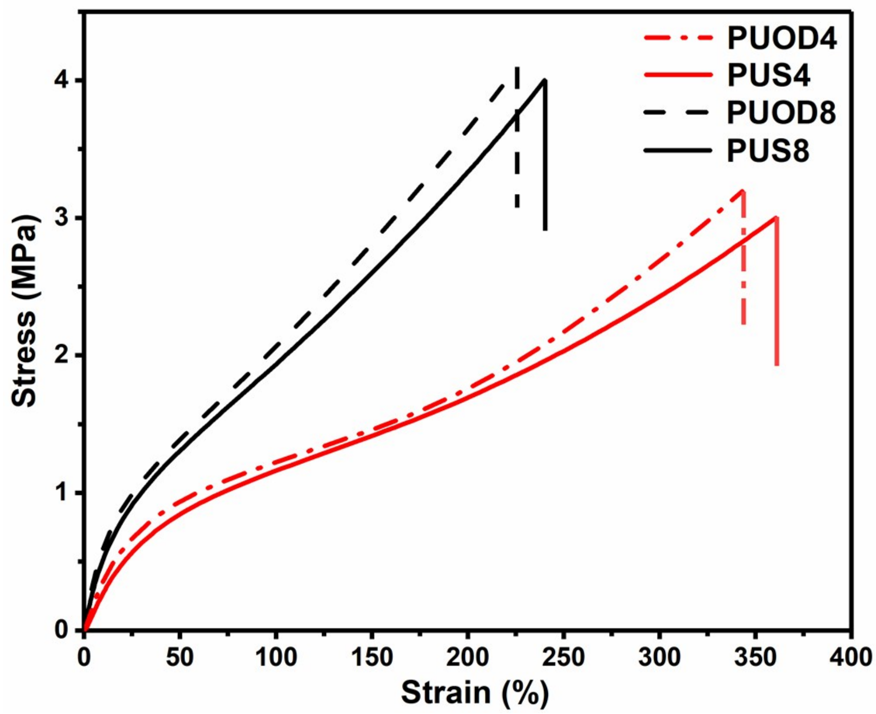

Figure S4. The stress-strain curves of the nanocomposite containing disulfide bonds and irreversible covalent bonds.

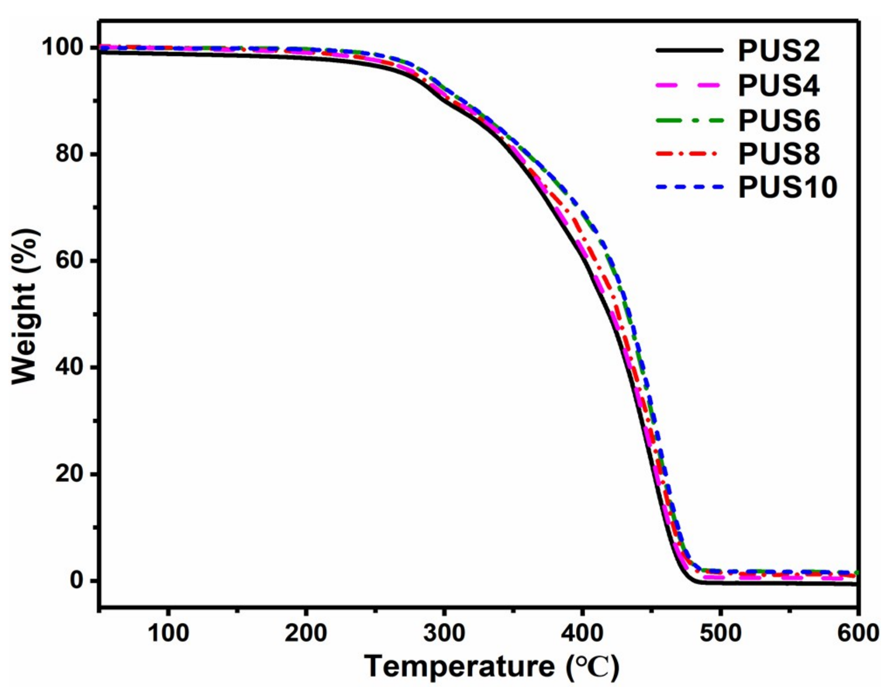

Figure S5. TGA curves of flexible U-MWC-PUS nanocomposite.

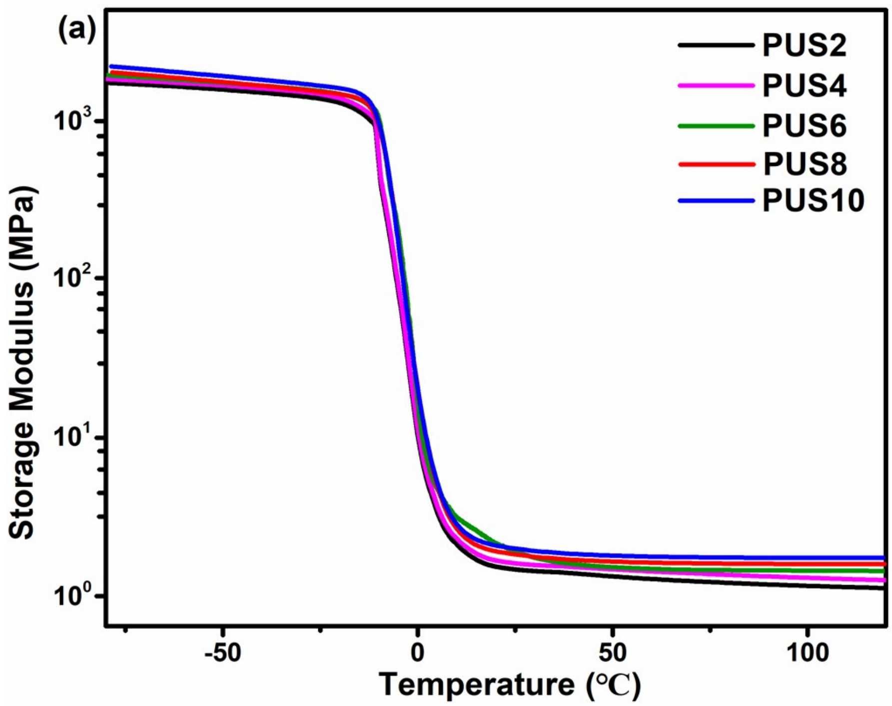

Figure S6. Storage modulus of flexible U-MWC-PUS nanocomposite.

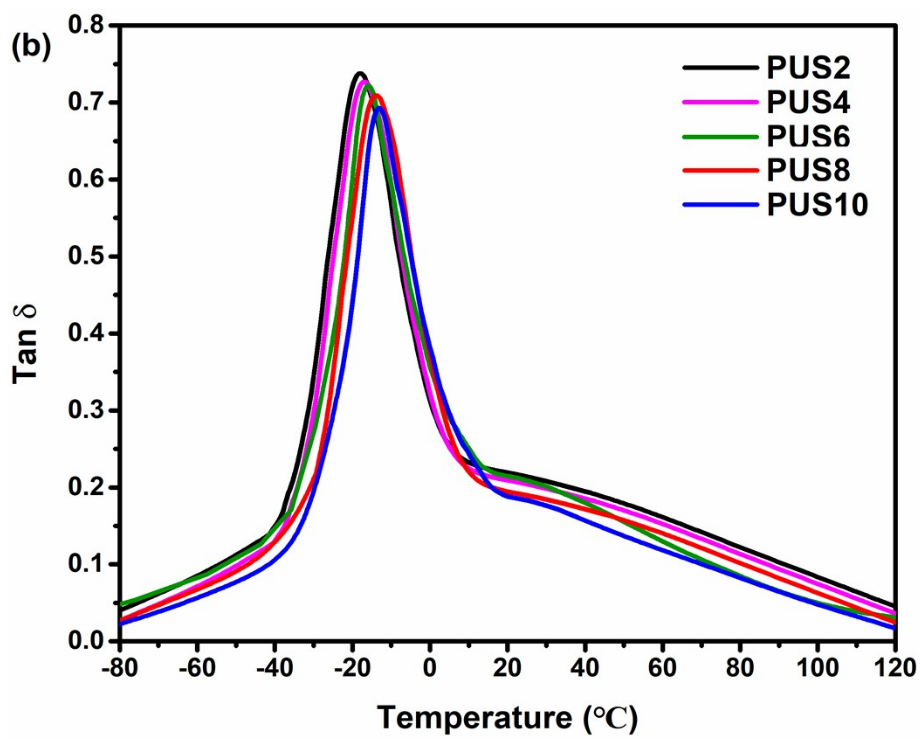

Figure S7. Tan δ curves of flexible U-MWC-PUS nanocomposite.

Table S3. Mechanical properties of PUS6, PUS6* and PUOD6

Sample	Original		Self-healed		Self-healing efficeiency (%)
	Tensile strength (MPa)	Breaking strain (%)	Tensile strength (MPa)	Breaking strain (%)	
PUS6	3.58±0.06	258±9	3.29±0.12	234±15	92.0
PUS6*	3.15±0.05	270±10	2.67±0.08	221±11	84.7
PUOD6	3.65±0.08	243±11	0.34±0.05	15±4	9.2

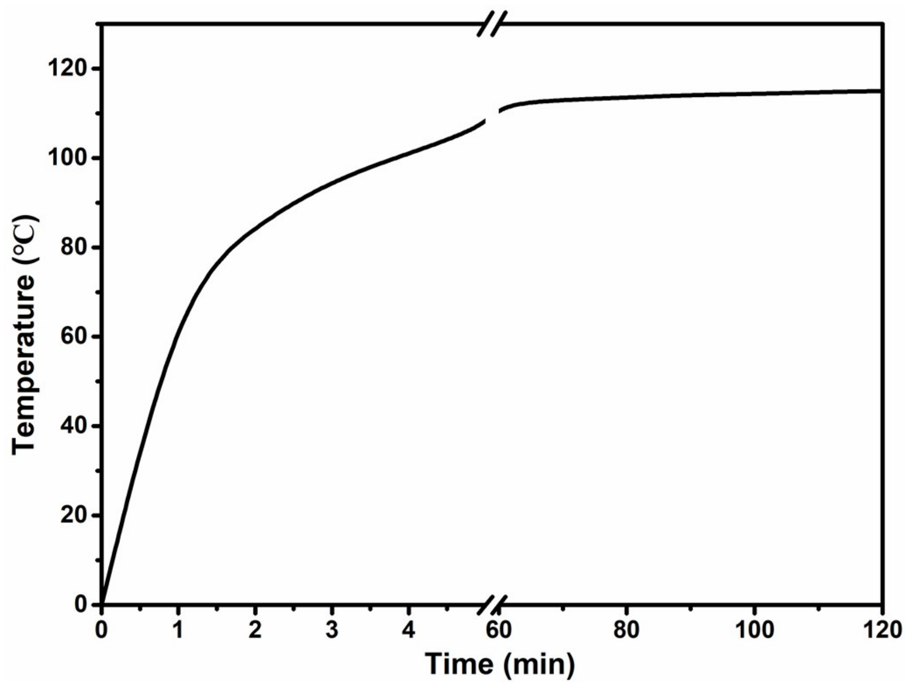

Figure S8. The temperature of the PUS6 crack surface varying with NIR irradiation time.

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