Supplementary Information (SI) for Journal of Materials Chemistry C.

Supporting Information

Self-healing epoxidized natural rubber flexible sensors based on hydrogen bonding interactions

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Testing and characterization

(1) Infrared spectral analysis (FTIR)

Infrared spectrograms were obtained by recording 4000-500 cm^{-1} wave numbers of MMT, UPy-NCO, MMT-UPy and MMT-UPy/ENR using the ATR unit in an infrared spectrometer (Thermo Scientific Nicolet iS5, USA). The number of scans was 32 with a resolution of 4 cm^{-1}.

(2) Scanning electron microscope (SEM)

A scanning electron microscope (TESCAN MIRA LMS, Czech Republic) was used to observe the cross-sectional morphology of the MMT- UPy/ENR samples. The test distance was 8 mm and the accelerating voltage was 5 kV. the samples were treated with liquid nitrogen embrittlement before testing and a conductive environment was provided by gold spraying.

(3) Polarized Light Microscope (PLM)

Polarized light microscope (PLM, LW35PB) was used to observe the macroscopic morphology of the MMT-UPy/ENR composite cross section. The magnifications were 50, 100, 400 and 600 times.

(4) Thermal weight loss analyzer (TG)

A heat loss weight analyzer (PerkinElmer 4000, China) was used to test ENR, MMT-UPy3%/ENR, and MMT- UPy10%/ENR, respectively. The equipment was energized with nitrogen during the test, the temperature range was set at 25~700°C, and the heating rate was set at 10°C/min.

(5) Mechanical properties test

A universal tensile testing machine (Instron 4465, China) was used to perform tensile testing on the MMT- UPy/ENR series samples. The tensile rate was set at 20 mm/min. The specimens were rectangular strips, the thickness of the experimental specimens was 2 mm, the width was 4 mm, and the length of the test section was 15 mm.

(6) Rheological performance test

A rheometer (MCR 102e, Austria) was used for viscoelasticity testing of ENR and MMT-UPy/ENR series samples. Testing temperature: 25°C, frequency test range 0.1-100 Hz.

(7) Self-repairability test

In order to perform the self-repair efficiency test, the specimens were first cut from the center position, followed by docking the cut specimens together to ensure that the docking was tight and without gaps. Then, the docked specimens were placed in Petri dishes and repaired at a constant 45°C

The repair was carried out in a constant 45°C environment for 12 h. After completion of the repair, the tensile properties of the original specimens and the self-repaired specimens were tested and the self-repair efficiency was calculated according to Equation (1):

$$\eta = \frac{Stress_{\mathrm{healed}}}{Stress_{\mathrm{original}}} \times 100\% \qquad (1)$$

Where: η-Self-repair efficiency

Stress healed-Tensile strength of the repaired sample;

Stress original-Tensile strength of the original sample.

(8) Infrared thermal imaging test

An infrared thermal imager (LT7-P, China) was used to plot the surface temperature of MMT-UPy/ENR and MMT-UPy/ENR/sandwiched NPC-MCNT composites with time. The samples were cut into circular slices of the same size, with a diameter of 1.5 cm and a thickness of 0.8 mm, and placed on a thermostatic heating table at 100°C for infrared thermography of the warming process, and then placed on the ground for infrared thermography of the cooling process when the temperature stabilized.

(9) conductivity test

A digital multimeter (Keithley 2450, USA) was used to test the electrical resistance of the MMT- UPy/ENR/Interlayer NPC-MCNT composites when they were placed horizontally at room temperature and pressure. The composites were cut into sample strips with dimensions of 5 cm × 1 cm × 0.8 mm and connected to the tester at both ends.

(10) Human Motion Performance Test

A digital multimeter (Keithley 2450, USA) was used for the human motion monitoring ability test. The conductive tape was inserted into both ends of the composite material during sample making, and the sample specimens of the human body sensing test were directly fixed in each test part of the human body. By testing the deformation of each part of the human body during movement, the change in electrical resistance of the sample adhered to the human body due to deformation can be detected, thus realizing accurate detection of human movement, and the change in electrical resistance is obtained by Equation (2):

$$\Delta R / R_0 = (R - R_0)/\mathrm{R}_0 \quad (2)$$

R_0: Resistance of the sample in the initial state

R : Resistance of the sample after deformation.

Table S1. Thermal decomposition temperature parameters of ENR and MMT-UPy/ENR composites

Sample name	Thermal decomposition onset temperature/°C	Thermal decomposition center temperature (first stage)/°C	Thermal decomposition center temperature (second stage)/°C	Thermal decompos termination temperature/°C
ENR	320	375	—	451
MMT-UPy$_{3\%}$/ENR	333	383	444	468
MMT-UPy$_{10\%}$/ENR	341	390	445	478

Table S2. Stress-strain data for MMT-UPy/ENR

Sample name	MMT-UPy$_{1\%}$/ENR	MMT-UPy$_{3\%}$/ENR	MMT-UPy$_{5\%}$/ENR	MMT-UPy$_{7\%}$/ENR	MMT-UPy$_{10\%}$/ENR
Stress(%)	794	957	885	985	1028
Strain(MPa)	0.81	0.89	1.02	1.13	1.27

Table S3. Stress-strain data of MMT-UPy/ENR after healing at 45℃, 12h

	Healing condition: atmospheric pressure, 45℃, 12h				
Sample name	MMT-UPy$_{1\%}$/ENR	MMT-UPy$_{3\%}$/ENR	MMT-UPy$_{5\%}$/ENR	MMT-UPy$_{7\%}$/ENR	MMT-UPy$_{10\%}$/ENR
Stress(%)	719	851	753	797	812
Strain(MPa)	0.734	0.792	0.861	0.935	1.028
Recovering efficiency(%)	90.60%	89.00%	84.45%	82.78%	81.01%

Table S4. A comparative analysis of the key performance metrics between our research endeavor and the recently published flexible, stretchable strain sensors in the field.

Materials	Stresses (MPa)	Strains (%)	Response time(ms)	Gauge factor		Cycles (under strain)	Ref.
				Stretch ability (%)	GF		
NR/N-BP/NR	≈0.8	600	21	0-50	108	2000(100%)	1
				50-250	98		
				250-500	669		
				500-520	2280		
GNPs/MWCNTs/SR	\	\	46	0-40	6.3	2000 (70%)	2
				40-90	40.1		
				90-100	2675.5		
XSBR/SSCNT	12.58	217	200	0-3	2.41	200(5%)	3
				0-170	4.24		
				170-214	25.98		
CPFC	\	\	400	0-17.6	7.19	2400(90%)	4
				21-180	2.63		
S-MXenes/S-ENR	4.55±0.12	388.33 ±30.52	50	\	107.43	5000(10%)	5
PDA/NBR/CB	≈9	540		0-180	346	1000（20%）	6
$CNT_{0.23}$/PEDOT:$PSS_{0.5}$@NR	2.05	1878	63	\	\	2000（100%\2000%）	7
CF/CB-$RTV_{6\%}$	2.6	700	\	0-375	110	300000（160%）	8
				375-615	375-615		
				615-700	182		
MXene/CNTs/fluoro-rubber	\	\	21.2	1-50	110	1000（150%）	9
				50-80	231		
				80-100	431		
MMT-UPy/ENR/Sandwiched NCP-MCNT	1.27	1028	149.41	0-10	1.75	800(50%)	Our work
				10-75	1.31		
				75-100	0.88		

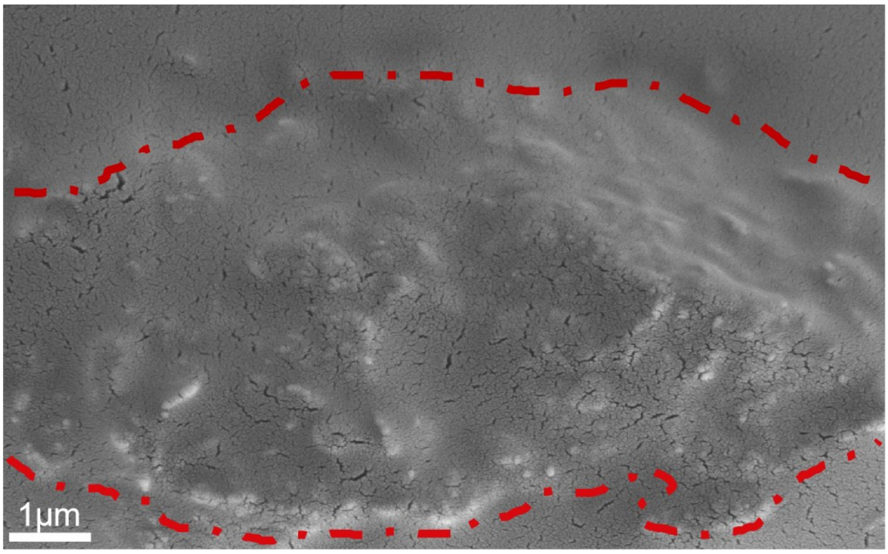

Fig.S1 Schematic of sandwich structure of MMT-UPy/ENR/Sandwiched NCP-MWCNT

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