

Supporting Information

Photothermal-responsive self-healing PDMS-based elastomers for stretchable strain sensors

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Materials

Amino-capped polydimethylsiloxane (PDMS, Mw: 3000) was purchased from Shanghai Macklin Biochemical Co., Ltd; isophorone diisocyanate (IPDI, 99%), 4, 4'-Dithiodianiline(DTSA, 98%) was bought from Anhui Senrise Technologies Co., Ltd; 2, 4, 6, 8-tetramethyl-2, 4, 6, 8-tetrakis[3-(oxiranylethoxy) propyl] cyclotetrasiloxan (TEC, 97%) were provided from Zhengzhou Alfa Chemical Co., Ltd; tetrahydrofuran (THF), silver nanowires (AgNWs), and Dibutyltin dilaurate were provided from Shanghai Aladdin Biochemical Technology Co., Ltd; all of the reagents can be used without purification.

Preparation of PUSE elastomer

First, H₂N-PDMS-NH₂ (1.9, 1.8, and 1.7 mmol) was separately weighed into 20 mL sample vials and diluted with an appropriate amount of THF (15 mL). IPDI (4 mmol) was weighed and mixed with THF (15 mL). APDS (0.1, 0.2, and 0.3 mmol) was separately weighed and diluted with THF (5 mL). In addition, TEC (0.1 mmol) with a photoinitiator (5 uL, Triaryl selenonium hexafluoroantimonate solution 50wt% propylene carbonate solution) were diluted with THF (5 mL). Then, Under N₂ protection, the THF -diluted solutions of H₂N-PDMS-NH₂, IPDI, APDS and TEC were injected into a two-neck round-bottom flask using disposable syringes. The reaction mixture was heated to 60 °C and allowed to react for 24 h. Afterward, THF was removed, and the reaction was stirred at 60 °C for additional 6 h. Subsequently, the resulting reaction sol was poured into a 5 cm × 5 cm polytetrafluoroethylene (PTFE) mold and irradiated with a 365 nm UV lamp (80 W) for 2 h. After the elastomer surface became dry enough, the sample was transferred to a 60 °C oven and dried for 24 h, yielding the PUS₁E, PUS₂E, PUS₃E elastomers (with different APDS content of 0.1, 0.2, and 0.3 mmol), respectively. And PUE_X elastomers with different TEC content (X=0.1, 0.2, 0.3, 0.4, 0.5 mmol) and 2 mmol PDMS, were prepared by the same synthesis route and procedures as before, and PUE₁, PUE₂, PUE₃, PUE₄, PUE₅ were obtained, respectively.

Preparation of PUSE/AgNWs elastomer

First, AgNWs were roll-coated onto a 4×7 cm glass substrate to obtain an AgNWs film with a sheet resistance below 10 Ω /sq. The roll-coated AgNWs film was then dried in an oven at 160 °C for 30 minutes. This step not only removed the excess ethanol dispersant but also helped to form a continuous silver nanowire network, thereby reducing the resistance. Next, PUSE sol was drop-coated onto the AgNWs film and cured under 365 nm, 80 W UV light in a vacuum oven. After the elastomer cured and formed a film, it was peeled off from the glass to obtain the PUSE/AgNWs composite conductive film. Copper tape was applied to both sides of the composite conductive film to form a simple strain sensor for subsequent testing.

Characterization method

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR (Thermo Fisher NICOLET 6700) was used to characterize PDMS, IPDI, TEC, DTSA, PU, PUE and PUSE solution.

Thermodynamic Analysis (TGA, DSC)

TGA analysis was conducted using a TG209F1 instrument, where the sample was heated from 30 °C to 800 °C at a ramp rate of 10 °C min⁻¹ under an N₂ atmosphere. DSC analysis was performed using a DSC214 instrument, where the temperature range was set from -150 °C to 100 °C at a ramp rate of 10 °C min⁻¹ under an N₂ atmosphere.

Optical Micrograph

Images were captured using an industrial electron microscope TD-TZG5X-KHT instrument, at a magnification of 10x.

Surface Morphology Analysis

Surface roughness analysis was conducted using a scanning probe microscope (model: 3100 SPM). Surface topography analysis was performed using a cold-field high-resolution scanning electron microscope (SEM).

Tensile Test

The tensile test was conducted in accordance with GB/T 528-2009 (China) using

a 1 kN universal material testing machine. The PU, PUE, PUSE elastomer was prepared as a dumbbell-shaped sample with a length of 50 mm, width of 4 mm, and thickness of 2 mm. A tensile rate of 10 mm/min was applied. For tensile cycling tests, the sample was stretched to 10% and 100% strain at a rate of 10 mm/min, then returned to its initial position at the same rate of 10 mm/min.

Self-Healing Test

Surface scratch experiments were conducted on the PUE, PUSE elastomer using a utility knife. After healing under various conditions, the recovery of the surface scratches was observed under an optical microscope. The original PUE, PUSE dumbbell-shaped sample was cut in half at the center, and the sample was healed under different conditions: room temperature, a 70 °C oven, and 365 nm, 80W UV light for 24 hours. Subsequently, the tensile strength of the healed samples was tested using the universal material testing machine, and the self-healing efficiency of the elastomer was calculated by the ratio of the maximum stress of the healed sample to the initial sample.

Electric Heating Test

The PUSE/AgNWs conductive elastomer was subjected to an external voltage using a DC power supply. The surface temperature changes of the elastomer within a voltage range of 0.5 V to 3 V were measured using an infrared thermometer.

Human Motion Signal Monitoring

Copper tape was applied to both ends of the PUSE/AgNWs conductive elastomer to form a simple strain sensor. This strain sensor was placed on the finger joints, wrist joints, and pulse, and the change in the resistance of the elastomer during various movements was tested using an electrochemical workstation.

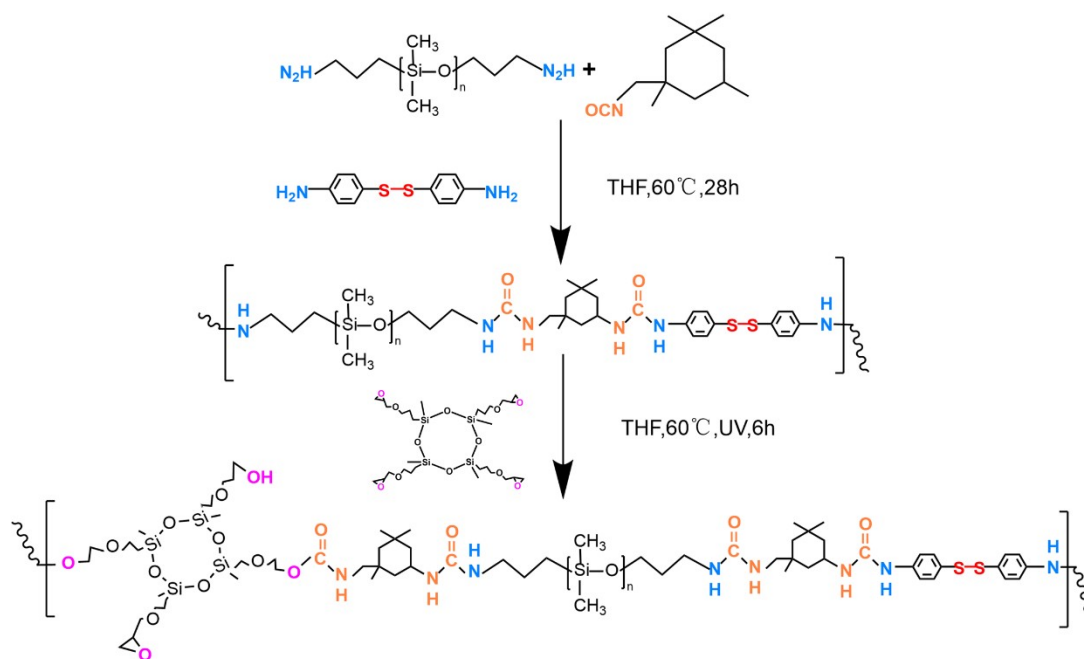


Figure S1. Synthesis route of PUSE.

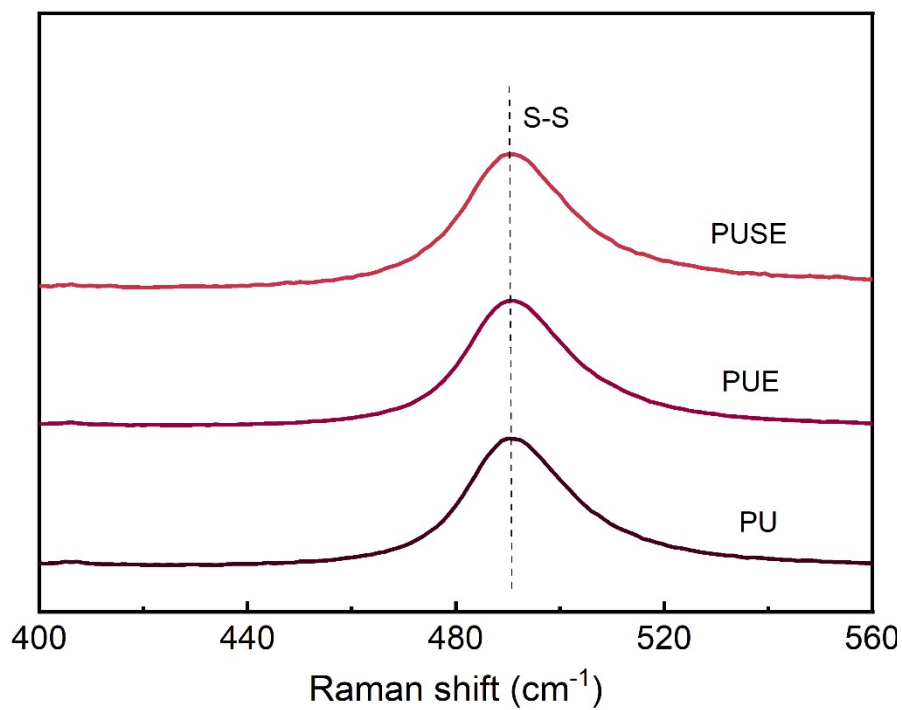


Figure S2. Raman spectrum of PUSE.

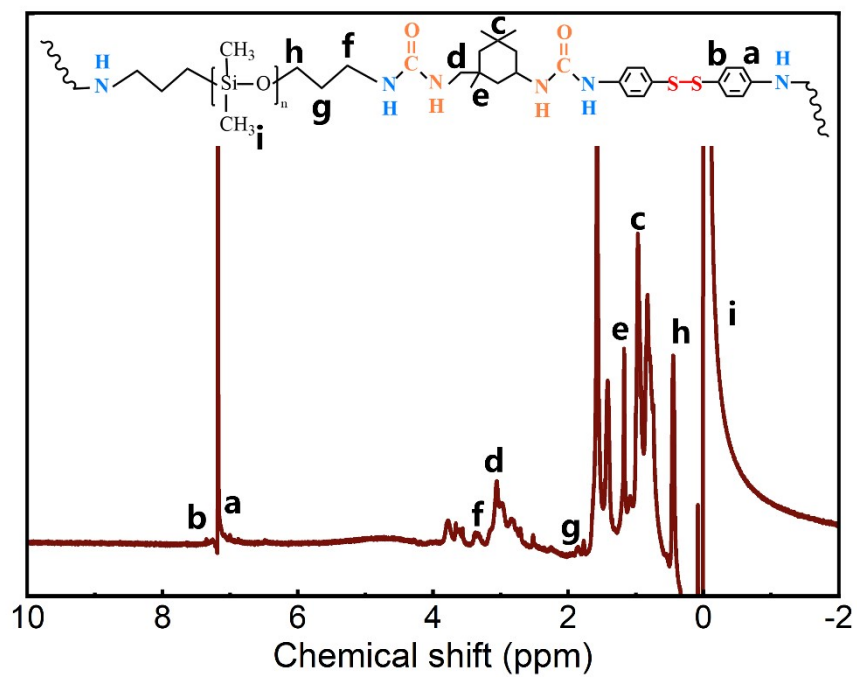


Figure S3. ¹H NMR spectrum of PUSE.

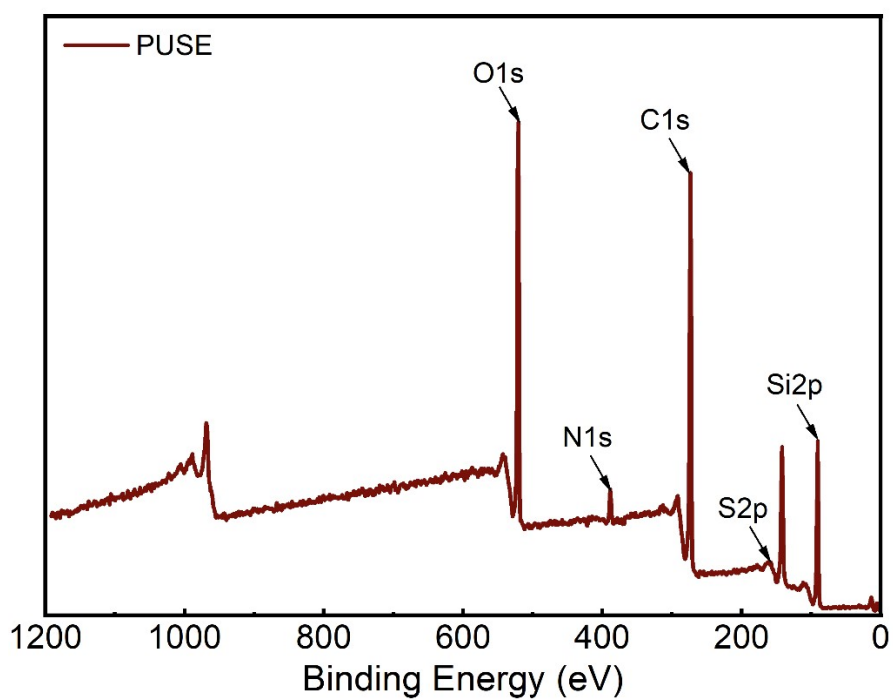


Figure S4. XPS spectrum of PUSE.

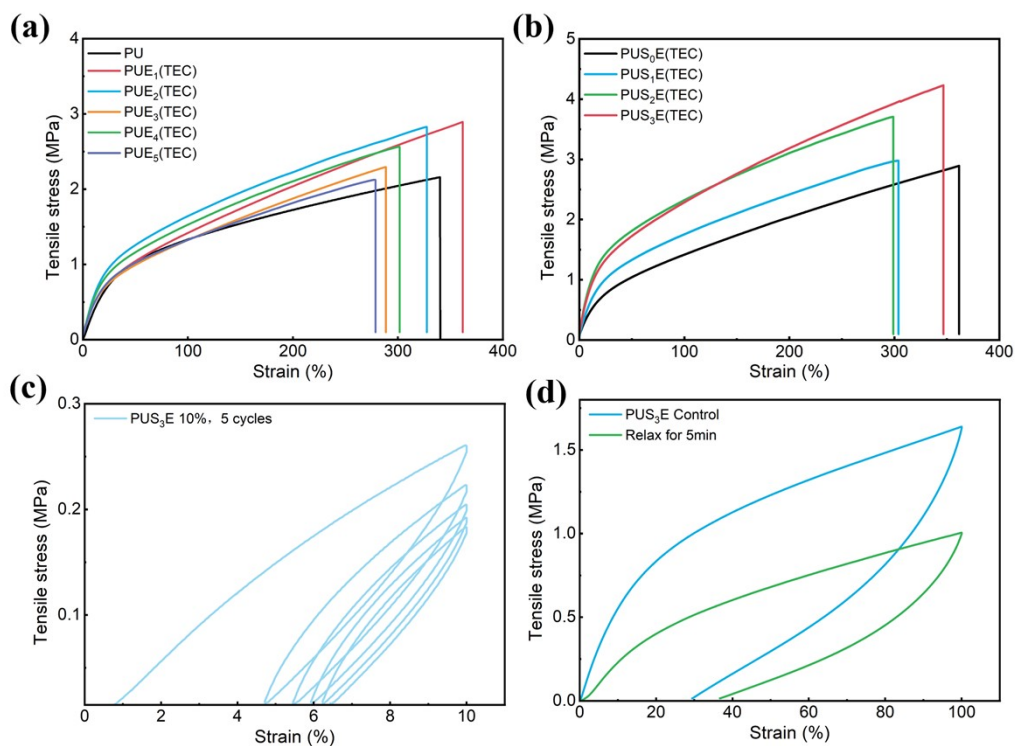


Figure S5. (a) Stress-strain curves of PUE_x elastomers with different TEC contents. (b) Stress-strain curves of PUS_xE elastomers with different APDS contents. (c) Cyclic loading-unloading tensile curves of the PUS_3E elastomer for 5 cycles at 10% strain. (d) Cyclic loading-unloading tensile curve of the PUS_3E elastomer after one cycle at 100% strain and relaxation for 5 minutes.

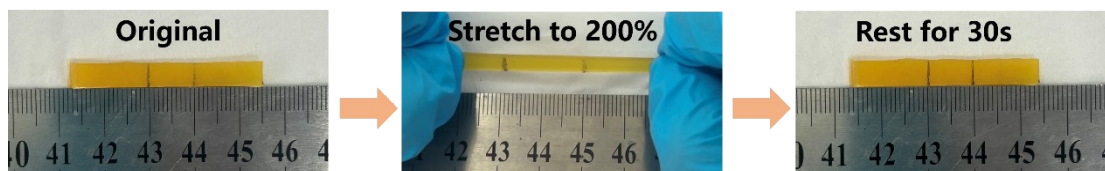


Figure S6. Photographs of the original, stretched and recovered PUS_3E .

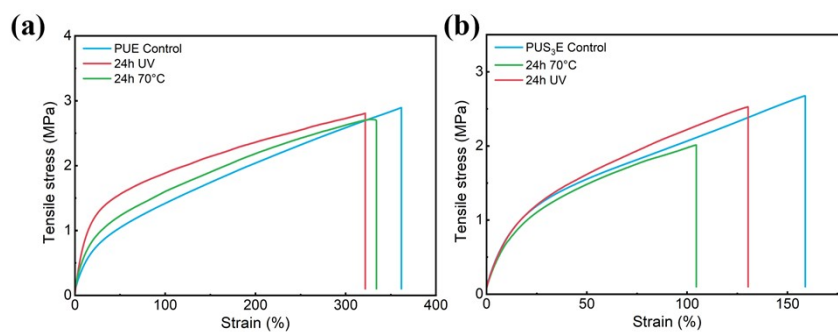


Figure S7. (a) Stress-strain curves of PUE elastomer before and after self-healing under different conditions (UV, 70°C). (b) Stress-strain curves of PUS_3E elastomer before and after self-healing under different conditions (UV, 70°C).

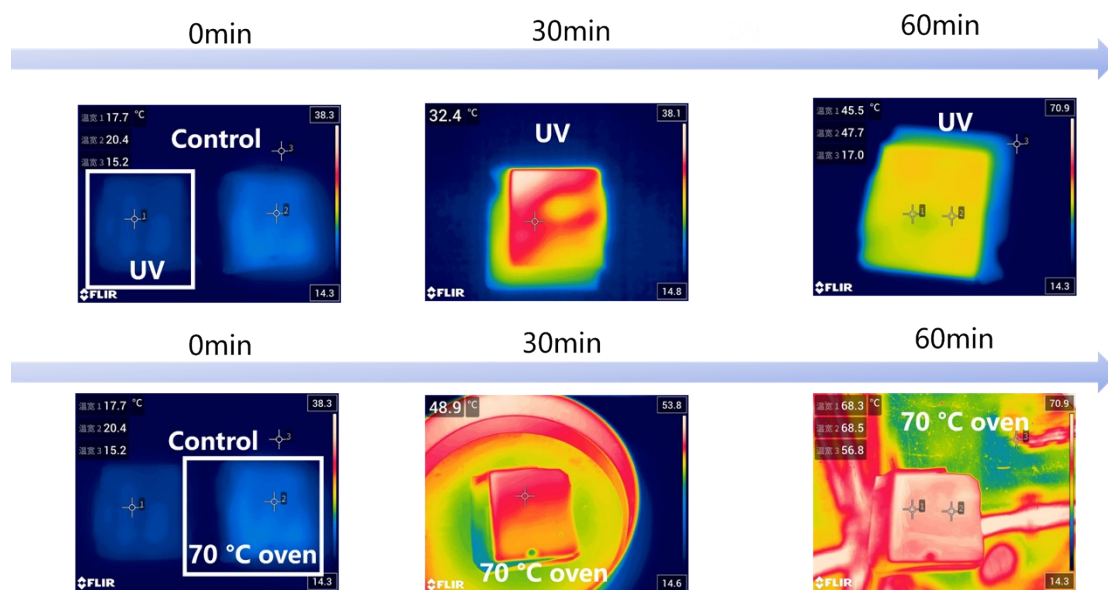


Figure S8. The infrared thermal images of the elastomer PUSE under UV irradiation and 70 °C heating conditions, respectively.

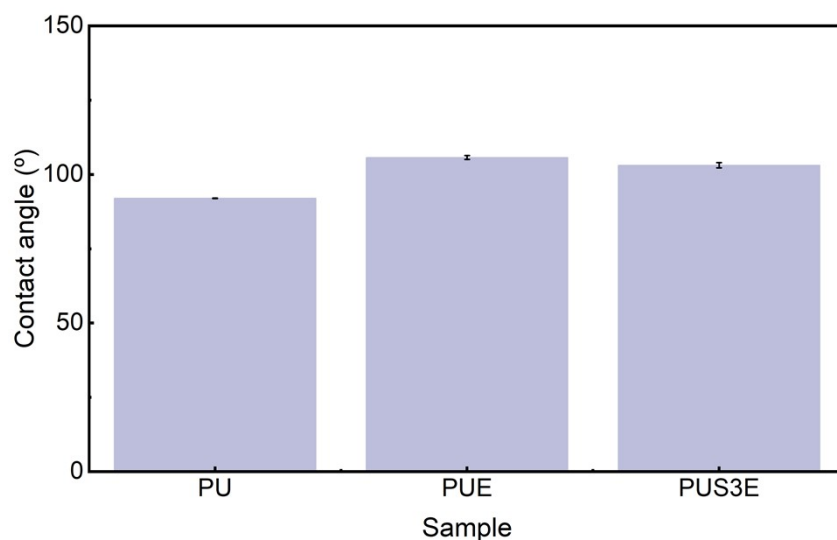


Figure S9. The water contact angles of PU, PUE, PUS₃E

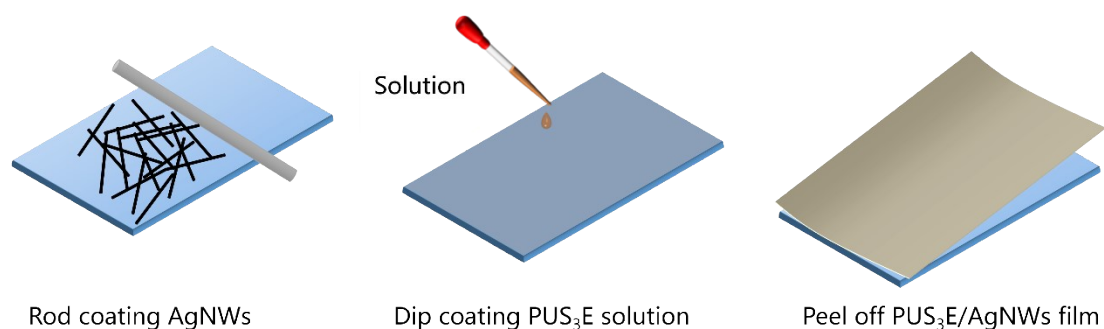


Figure S10. Schematic illustration of the fabrication process of the PUS₃E/AgNWs composite elastomer

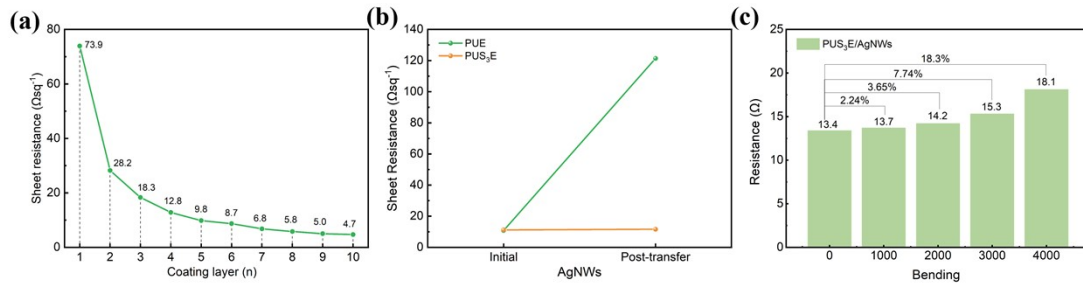


Figure S11. (a) Sheet resistance of AgNWs films with different coating layers. (b) Sheet resistance of AgNWs before and after compositing with PUE and PUS₃E elastomer. (c) Variation of sheet resistance of the PUS₃E/AgNWs composite conductive film before and after different bending cycles.

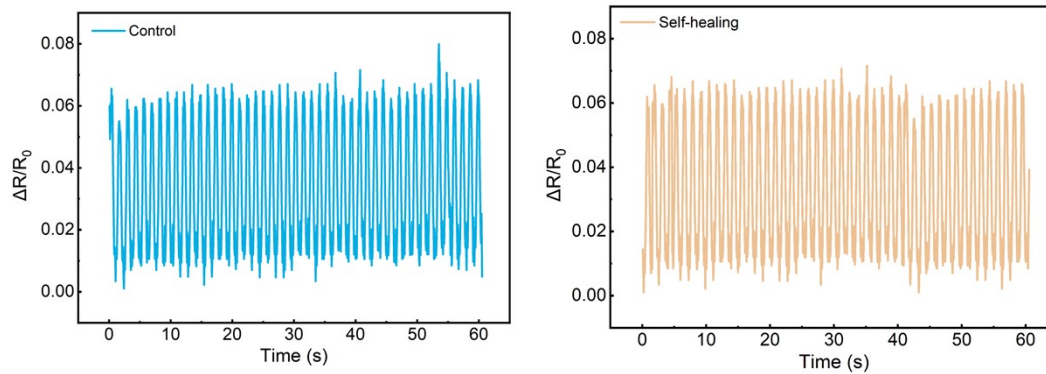


Figure S12. Pulse beat signal monitoring of PUS₃E/AgNWs strain sensors.

Table S1. Comparison of PUSE elastomers in the performance of material system, self-healing conditions, self-healing efficiency, tensile stress and toughness of this work with previously reported PDMS-based self-healing polymers.

Ref.	Material system	Self-healing conditions	Self-healing Efficiency	Tensile stress	Toughness
[1]	PDMS/Anthracene-based/photoresponsive segment	NIR light	90%	1.0 MPa	5.0 MJ·m ⁻³
[2]	PDMS/IPDI+disulfide/imine-containing chain extender	25 °C, 24 h	88%	1.5 MPa	6.0 MJ·m ⁻³
[3]	PDMS/Aromatic urea group	70 °C, 30 min	100%	0.87 MPa	2.1 MJ·m ⁻³
[4]	PDMS/Aldehyde-based crosslinker	25 °C, 24 h	75%	0.57 MPa	0.85 MJ·m ⁻³
[5]	PDMS/PVMA/aldehyde-modified carbon dots	UV/2 h	90%	0.9 MPa	6.5 MJ·m ⁻³
This work	PDMS/TEC/APDS	UV/24h	95%	4.2 MPa	9.85 MJ·m ⁻³

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