

Supplementary Information for
Autonomous Self-Healing Supramolecular Polymer Transistors for
Skin Electronics

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Supplementary Notes

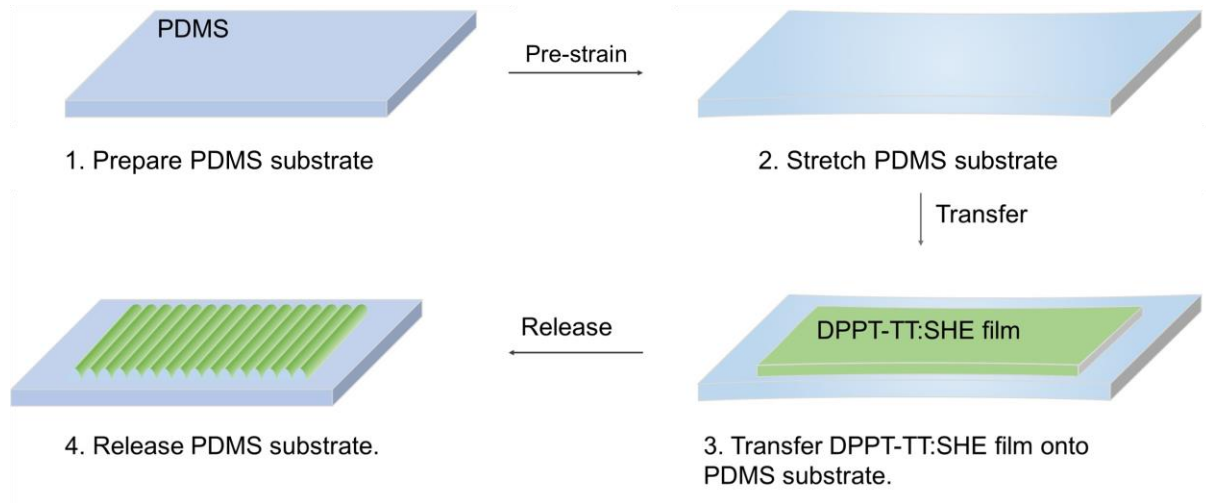
Supplementary Note 1. Characteristics of the FETs

The field effect mobility (μ_{FE}) is calculated in saturation regime by fitting the plot of the linear regime of square root of drain current ($\sqrt{I_D}$) versus gate voltage (V_G). The equation follows (S1):

$$\mu_{FE} = \frac{2L}{WC_i} \left(\frac{\partial \sqrt{I_D}}{\partial V_G} \right)^2 \quad (S1)$$

Where L is the length of the channel, W is width of the channel. C_i is capacitance per unit area of gate dielectric. The values of all parameters are shown at Table S1.

Supplementary Note 2. Calculation of elastic modulus using wrinkle forming method.



The elastic modulus of DPPT-TT:SHE films are measured using a buckling-based metrology¹. The DPPT-TT:SHE films are transferred onto pre-strained PDMS substrate. After releasing, the buckles of DPPT-TT:SHE film can be obtained. The elastic modulus of film is determined by equation (S2) and the optical images of buckles are shown Supplementary Fig. 4.

$$\frac{E_f}{(1-\nu_f^2)} = \frac{3E_s}{(1-\nu_s^2)} \left(\frac{d}{2\pi h_f} \right)^3 \quad (S2)$$

Where E_f is the Young's modulus of film, ν_f is the Poisson's ratio of the film, E_s is elastic modulus of substrate, ν_s is Poisson's ratio of substrate, d is wavelength of buckle and h_f is thickness of the film. In this determination, the elastic modulus of PDMS (E_s) is 2 MPa, Poisson ratio of PDMS (ν_s) is 0.5, Poisson ratio of the films (ν_f) is 0.35. The thickness of the films is obtained by ellipsometer.

Supplementary Note 3. Calculation method for dielectric constant (k)

The dielectric constant (Relative permittivity) is calculated following equation (S3).

$$k = \frac{C_i}{\epsilon_0} d \quad (S3)$$

Where k is dielectric constant, C_i is capacitance per unit area at 1 kHz, ϵ_0 is permittivity of vacuum and d is thickness of dielectric film. The structure and method for MIM structure are shown at Supplementary Fig. 29.

Supplementary Note 4. Calculation method for surface energies of the neat DPPT-TT, 3:7 blend film, and neat SHE elastomer

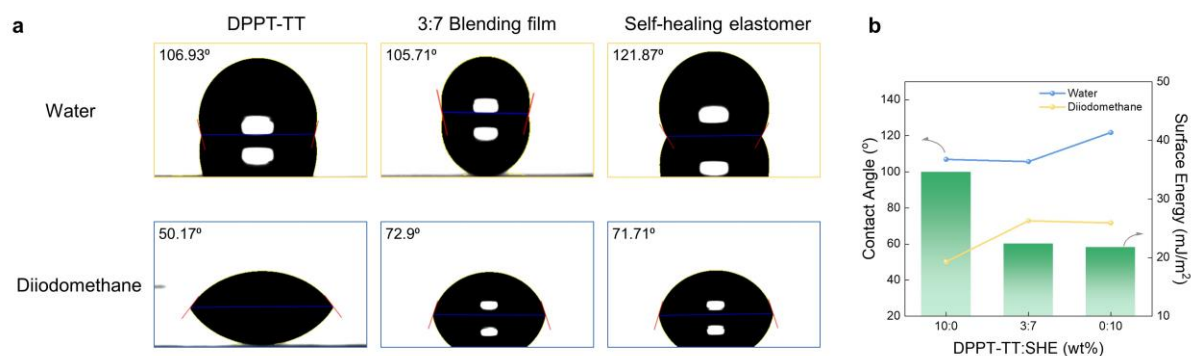
The surface free energies are calculated using the Owens-Wendt method²:

$$\gamma_c = \gamma_c^p + \gamma_c^d \quad (\text{S4})$$

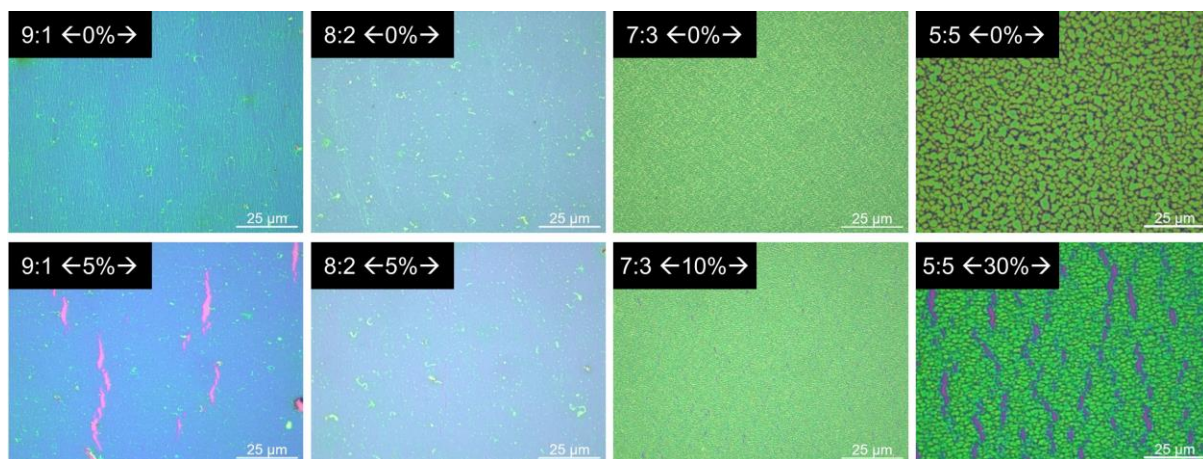
$$(1 + \cos \theta_l) \gamma_l = 2 \left(\sqrt{\gamma_l^d \gamma_c^d} + \sqrt{\gamma_l^p \gamma_c^p} \right) \quad (\text{S5})$$

where γ_c , γ_c^p , and γ_c^d are the total surface energy, polar component and dispersive component of surface energy of testing materials, respectively. Where θ_l , γ_l , γ_l^p , and γ_l^d are the contact angle, total surface energy, polar and dispersive component of surface energy of the test liquid, which are water and diiodomethane. $\gamma_{\text{diiodomethane}} = 50.8 \text{ mJ/m}^2$, $\gamma_{\text{water}} = 72.8 \text{ mJ/m}^2$, $\gamma_{\text{water}}^d = 21.8 \text{ mJ/m}^2$ and $\gamma_{\text{water}}^p = 51 \text{ mJ/m}^2$.

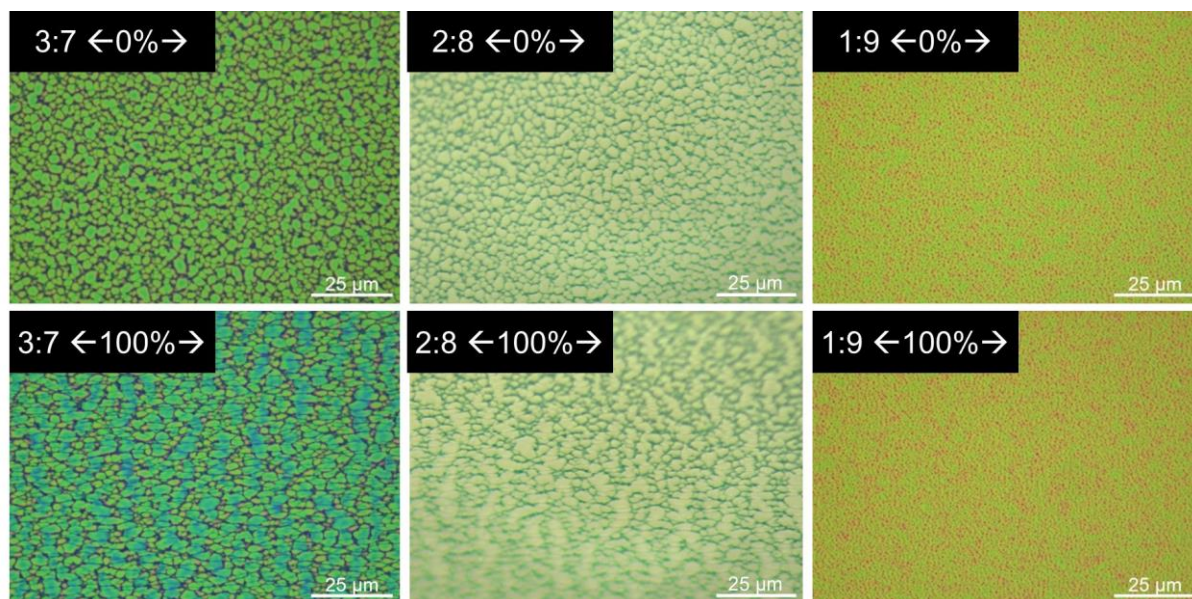
Supplementary Figures



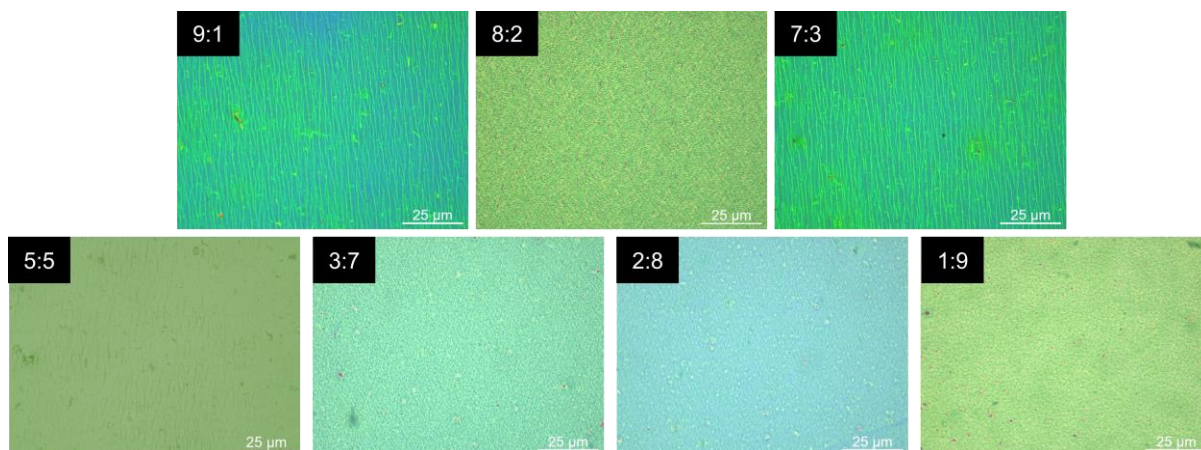
Supplementary Fig. 1. a, Optical images of the contact angles and **b**, surface energies of the neat DPPT-TT, 3:7 blending film and neat self-healing elastomer. The surface energies of neat DPPT-TT, DPPT-TT:SHE (3:7 weight ratio), and neat SHE are 34.55 mJ/m², 22.32 mJ/m², 21.7 mJ/m², respectively.



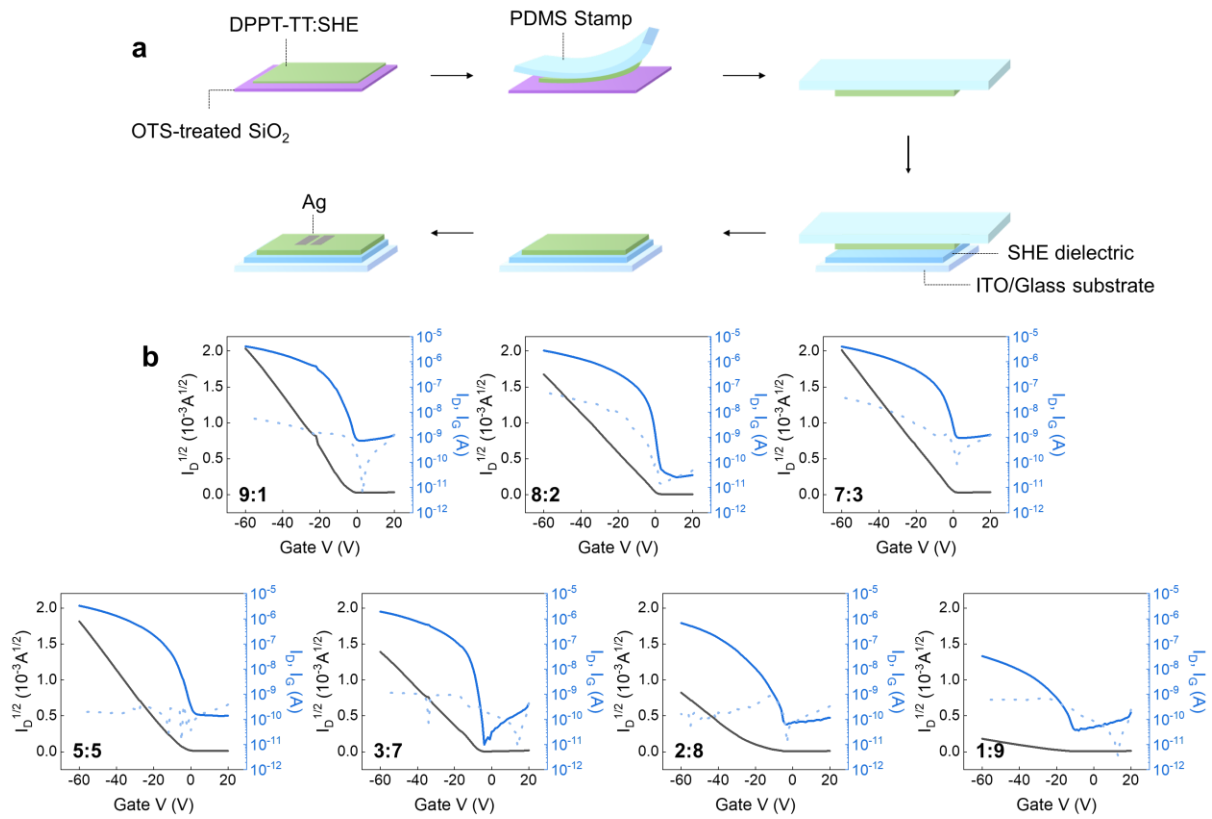
Supplementary Fig. 2. OM images at 0% (top) and crack on-set (bottom) strain of blend films (DPPT-TT:SHE) from 9:1 to 5:5 weight ratio.



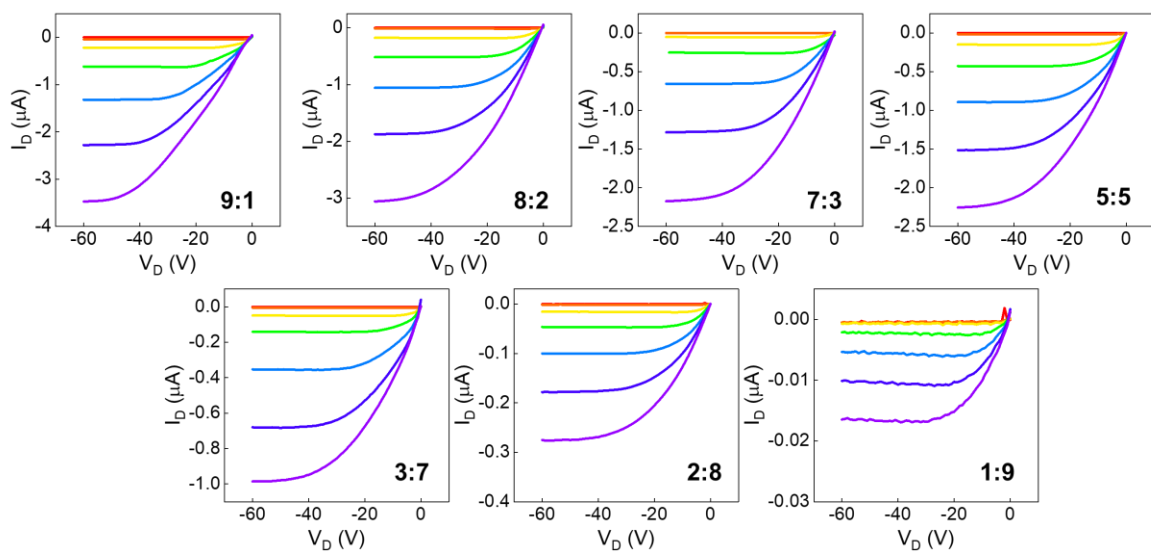
Supplementary Fig. 3. OM images at 0% (top) and 100% (bottom) strain of blend films (DPPT-TT:SHE) with 3:7, 2:8, and 1:9 ratio.



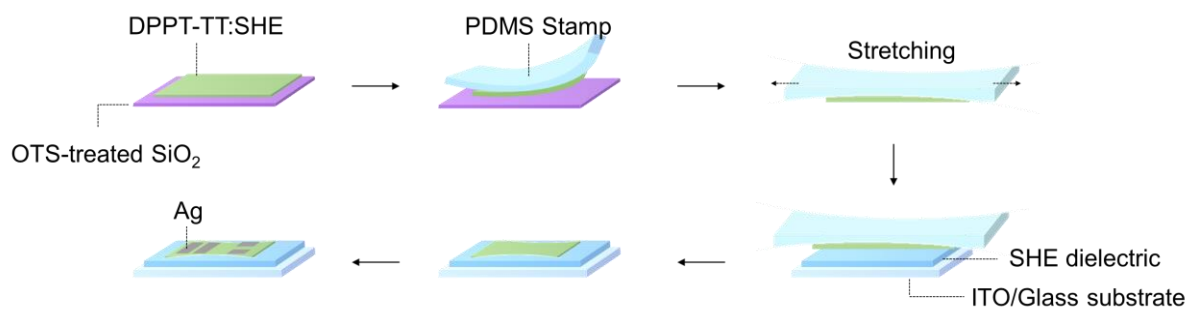
Supplementary Fig. 4. OM images of buckled blend films (DPPT-TT:SHE) with various blending ratios for elastic modulus. The elastic modulus of the films is calculated with equation S2 and Supplementary Note 2. The thickness of all blending films is approximately 100 nm.



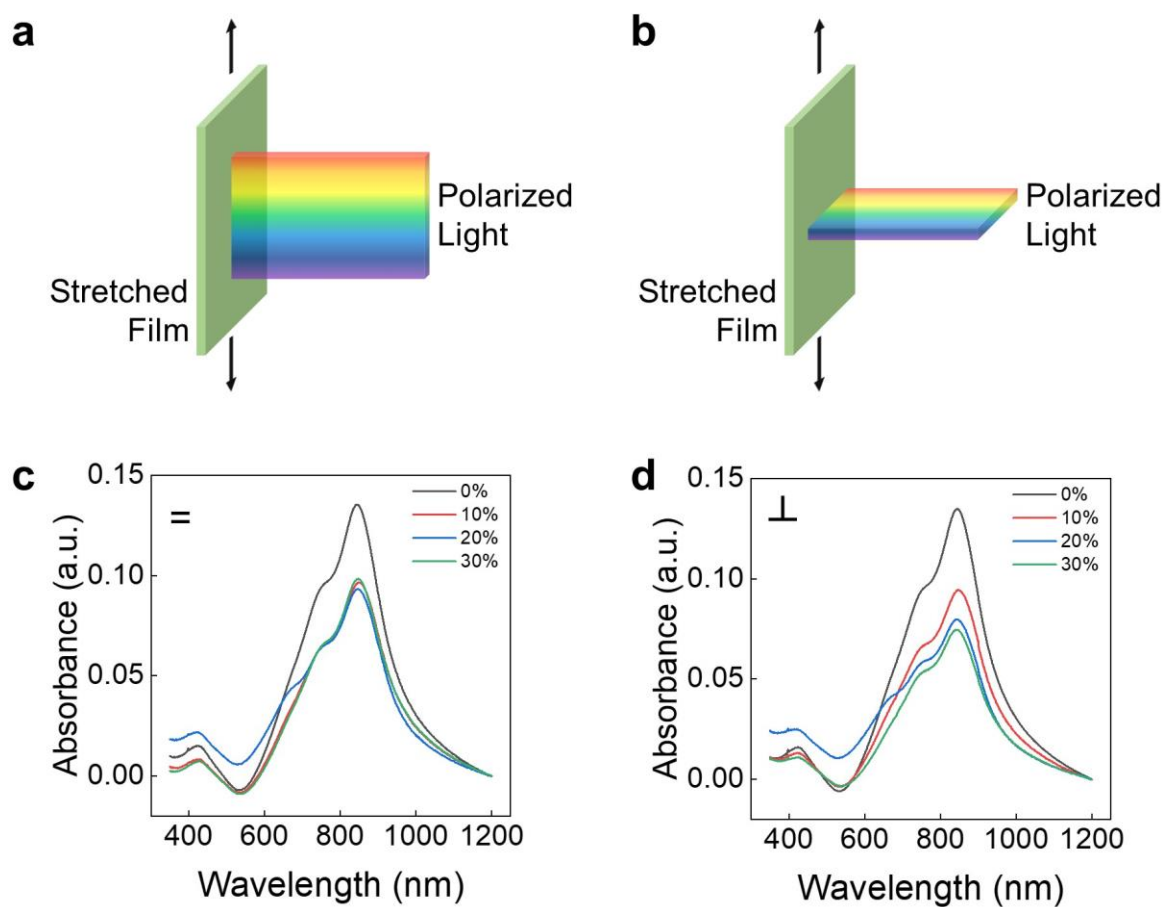
Supplementary Fig. 5. a, Transistor structure that is fabricated on rigid substrate (ITO-glass). **b**, The transfer characteristics of the transistors as a function of blend ratios (DPPT-TT:SHE).



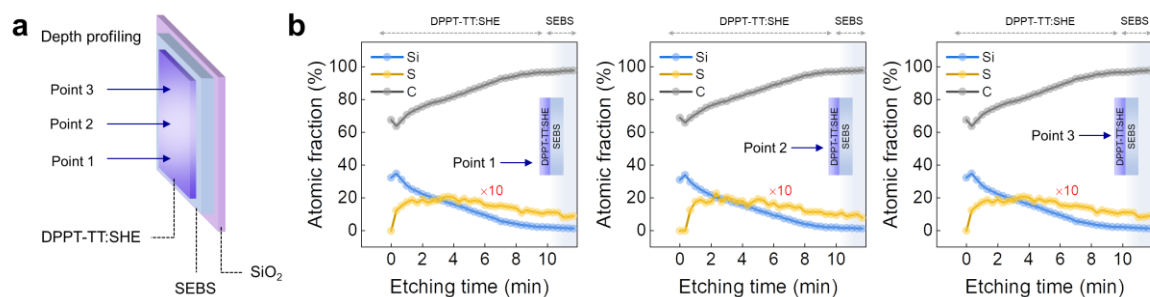
Supplementary Fig. 6. The output characteristics (V_G : 0 to -60 V, step: -10 V) of the blend films with various blend ratios (DPPT-TT:SHE).



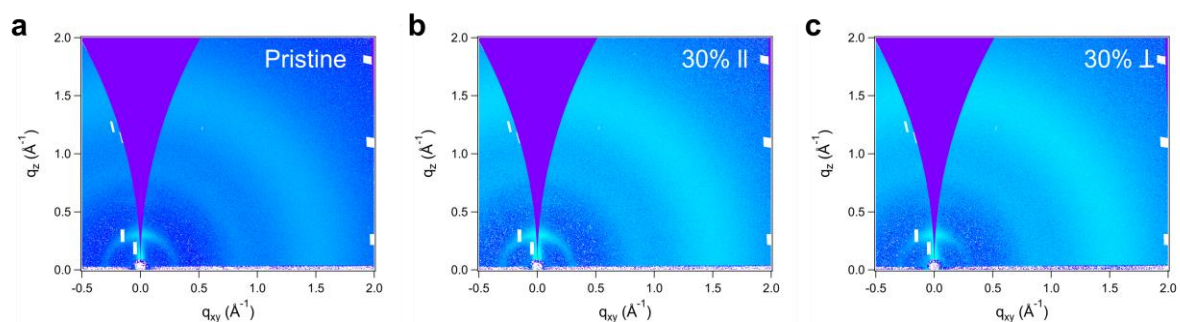
Supplementary Fig. 7. Schematic illustration of transistor structure for electrical measurement. The stretched-semiconducting film was transferred onto SEBS/ITO substrate, and the silver source and drain electrode was then deposited onto the semiconducting films.



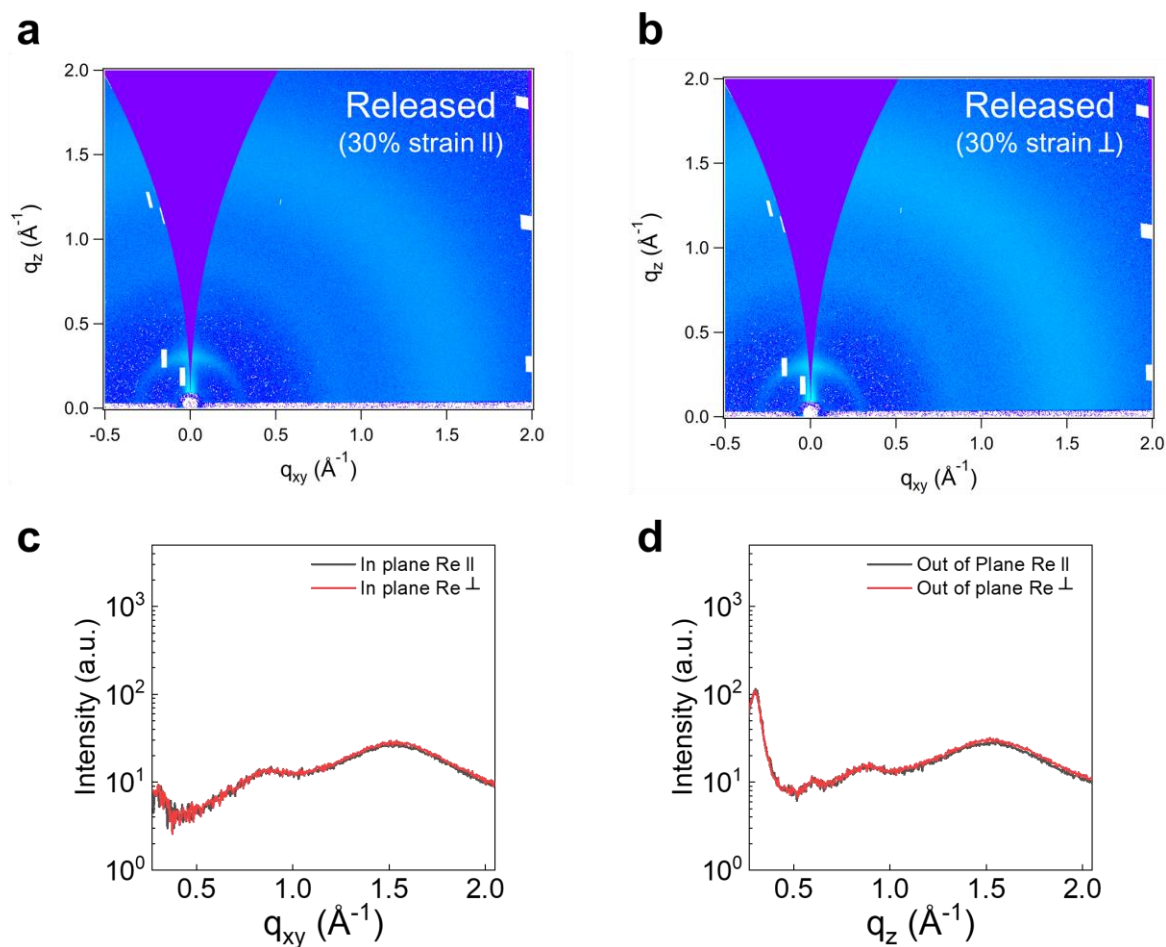
Supplementary Fig. 8. Schematic illustrations of polarized UV-vis-NIR characterization of the films. Polarized light in **a**, parallel and **b**, perpendicular directions to the stretching direction. Polarized UV-vis-NIR spectra of the blend film under 0% to 30% strain in **c**, parallel and **d**, perpendicular directions to the stretching direction.



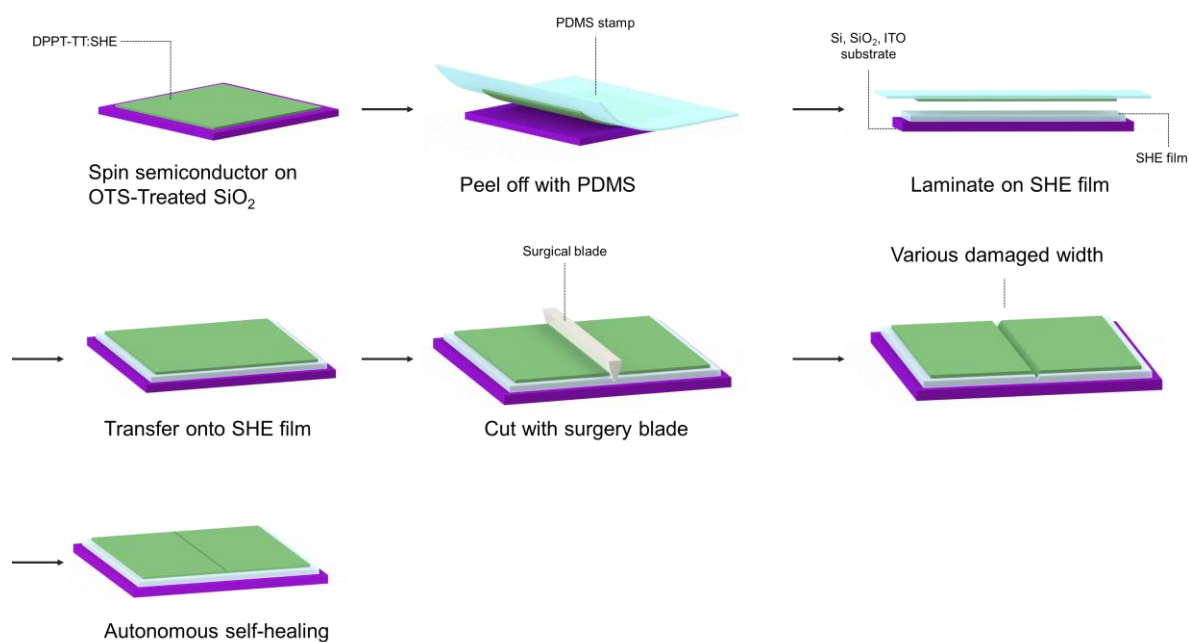
Supplementary Fig. 9. a, Schematic illustration of the sample structure for XPS analysis. The DPPT-TT:SHE blending film, in a 3:7 ratio, was spin-coated onto OTS-treated SiO₂. This blending film was then transferred onto an SEBS/SiO₂ substrate, where the SEBS layer served as a support for the separation of Si atoms from both DPPT-TT:SHE film and SiO₂ substrate. Notably, the SEBS consists only of C and H atoms. **b**, XPS depth profiling of atomic fraction in DPPT-TT:SHE films with three different point.



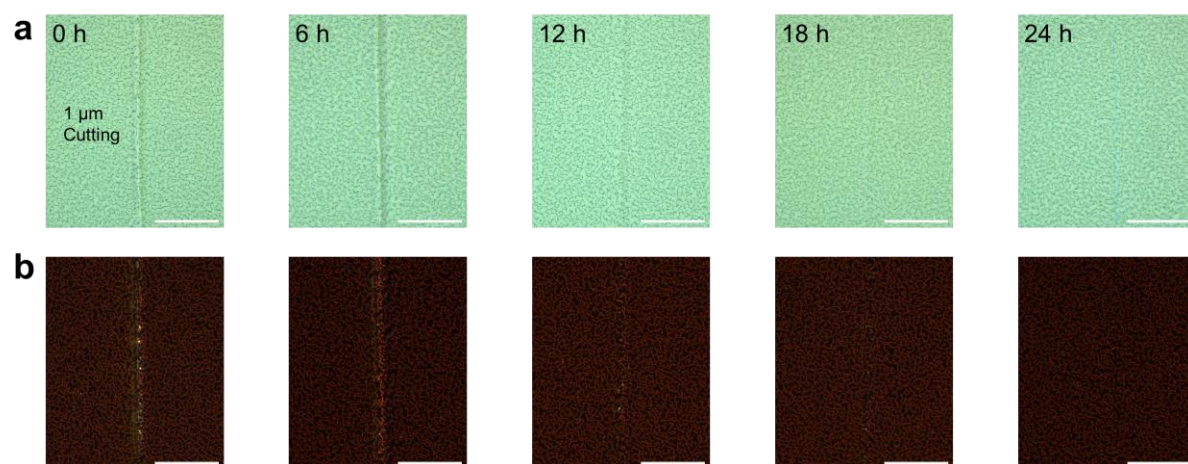
Supplementary Fig. 10. Two-dimensional GIXD patterns of the blend film (DPPT-TT:SHE=3:7 weight ratio) at **a**, pristine and stretched films under 30% strain in **b**, parallel, and **c**, perpendicular to the incident beam orientation.



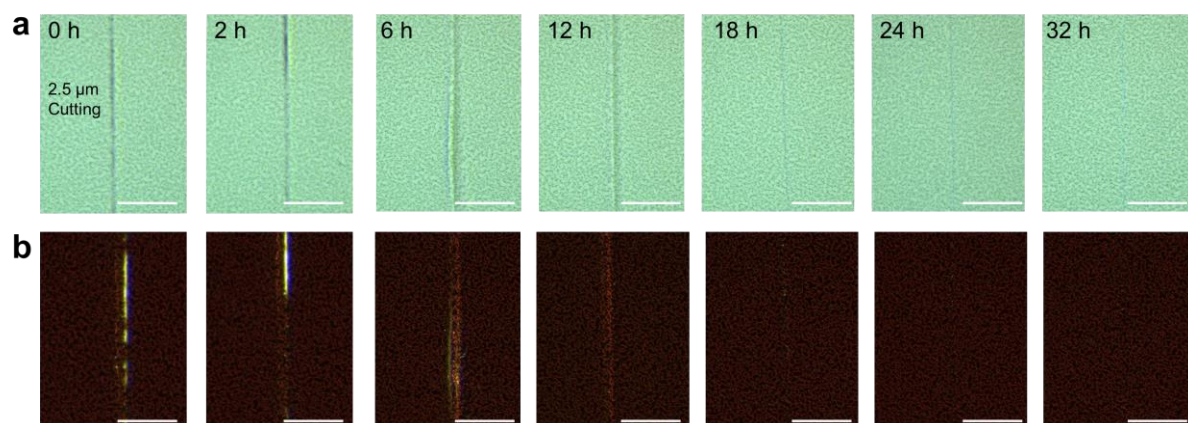
Supplementary Fig. 11. Two-dimensional GIXD patterns of the blend film (DPPT-TT:SHE=3:7 weight ratio) after stretching (30 % strain) in **a**, parallel and **b**, perpendicular to stretching-releasing direction. The intensity line cuts for the releasing film along **c**, in plane and **d**, out of plane of the films.



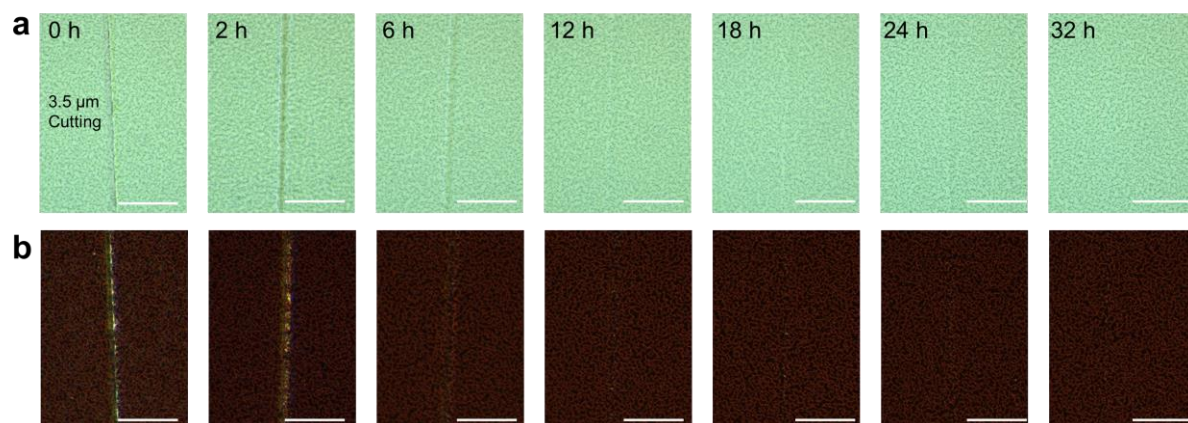
Supplementary Fig. 12. Schematic illustrations of autonomous self-healing process. The semiconducting film is transferred onto SHE film. We used various substrates such as Si, SiO_2 , and ITO-glass for device fabrication and materials analysis. The semiconducting film was cut using a surgery blade, resulted in microscale damage width up to $5\mu\text{m}$.



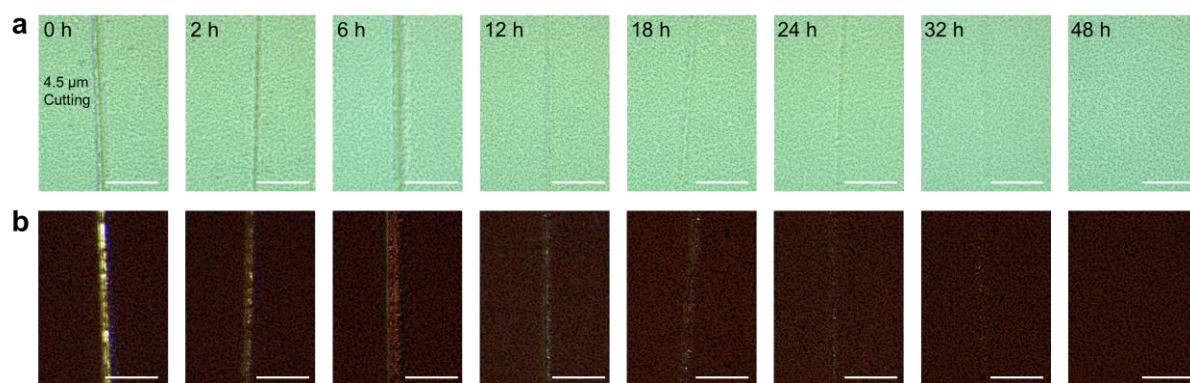
Supplementary Fig. 13. Optical images of about 1 μm damaged area with time flows at room temperature: **a**, bright field images and **b**, dark field images. Scale bar: 50 μm .



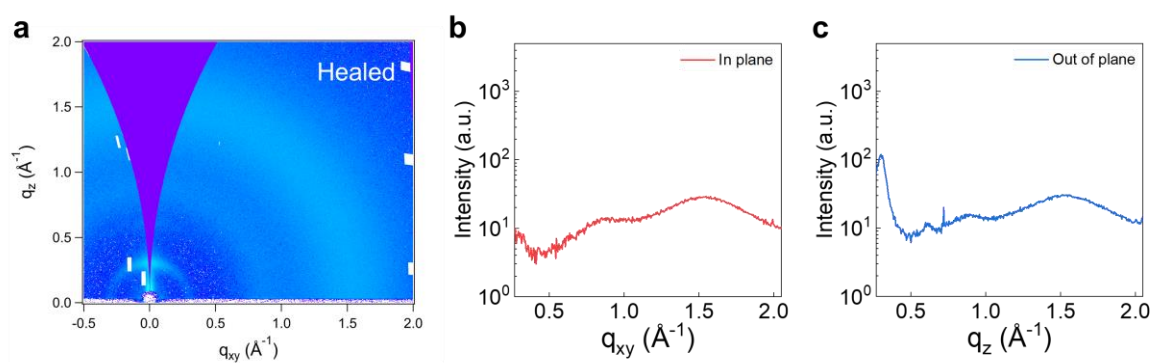
Supplementary Fig. 14. OM images of the damaged blend film (damage width: 2.5 μm) as a function of healing time at room temperature; **a**, bright field images and **b**, dark field images. Scale bar: 50 μm .



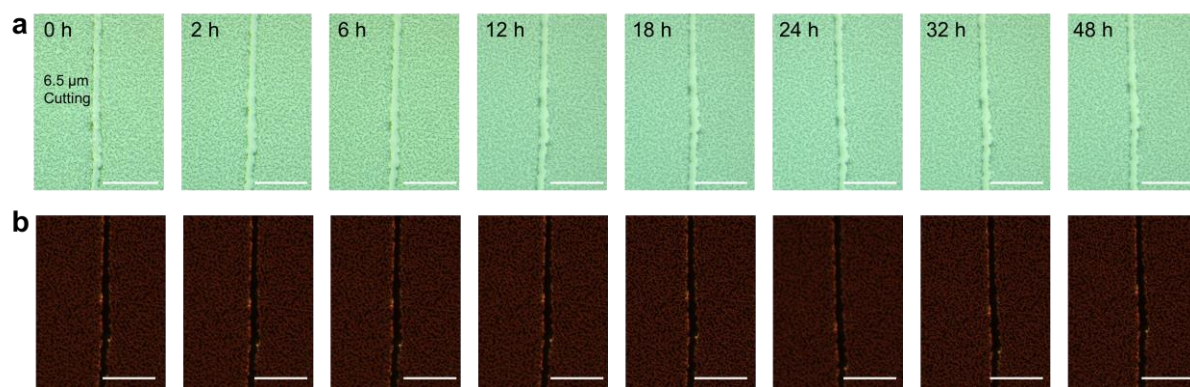
Supplementary Fig. 15. OM images of the damaged blend film (damage width: 3.5 μm) as a function of healing time at room temperature; **a**, bright field images and **b**, dark field images. Scale bar: 50 μm .



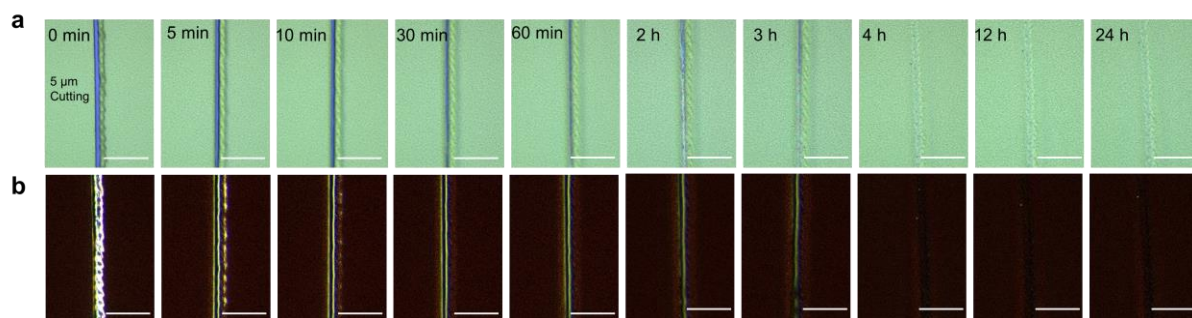
Supplementary Fig. 16. OM images of the damaged blend film (damage width: 4.5 μm) as a function of healing time at room temperature; **a**, bright field images and **b**, dark field images. Scale bar: 50 μm .



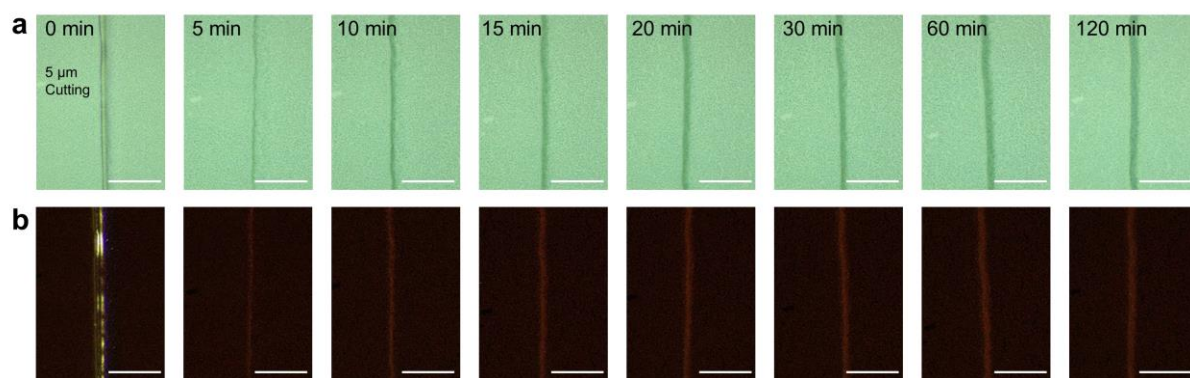
Supplementary Fig. 17. **a**, Two dimensional GIXD pattern of the healed semiconducting film. **b**, The intensity line cuts along in plane and **c**, out of plane of the healed film.



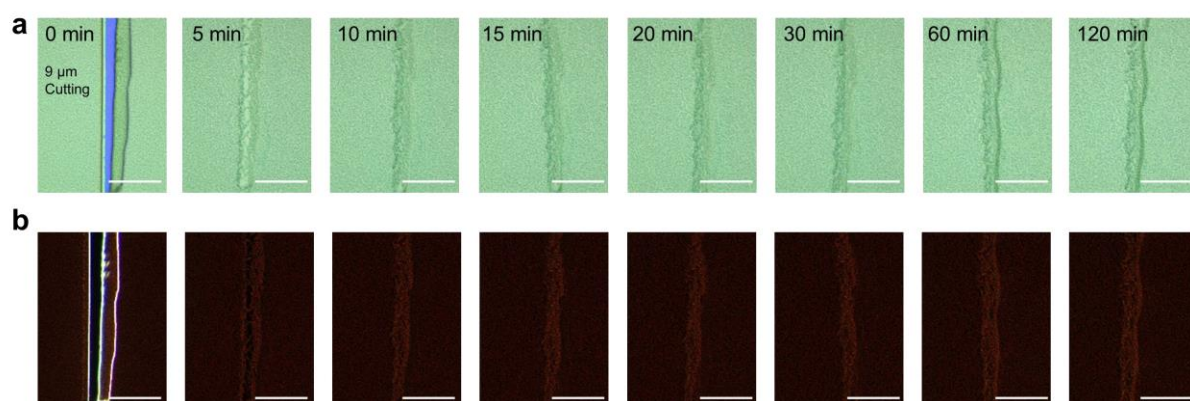
Supplementary Fig. 18. OM images of the damaged blend film (damage width: 6.5 μm) as a function of healing time at room temperature; **a**, bright field images and **b**, dark field images. Scale bar: 50 μm .



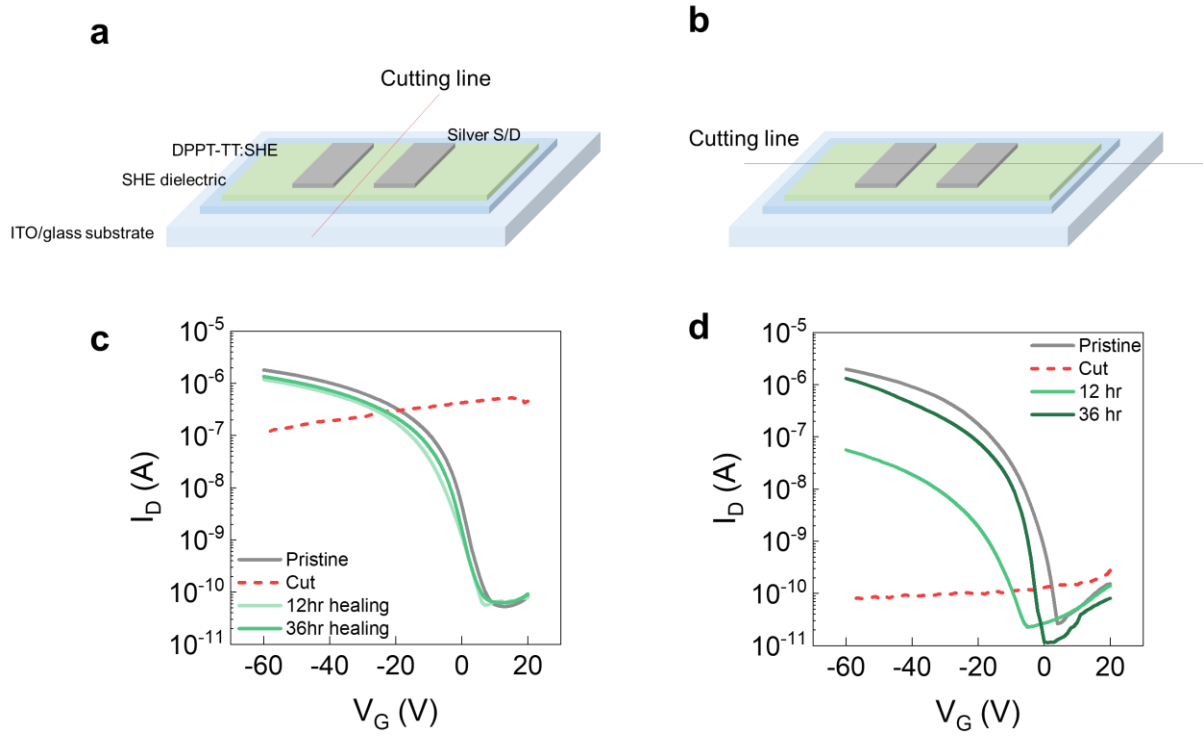
Supplementary Fig. 19. OM images of the damaged blend film (damage width: 5 μm) as a function of the time of heat treatment at 40 °C: **a**, bright field images and **b**, dark field images. Scale bar: 50 μm .



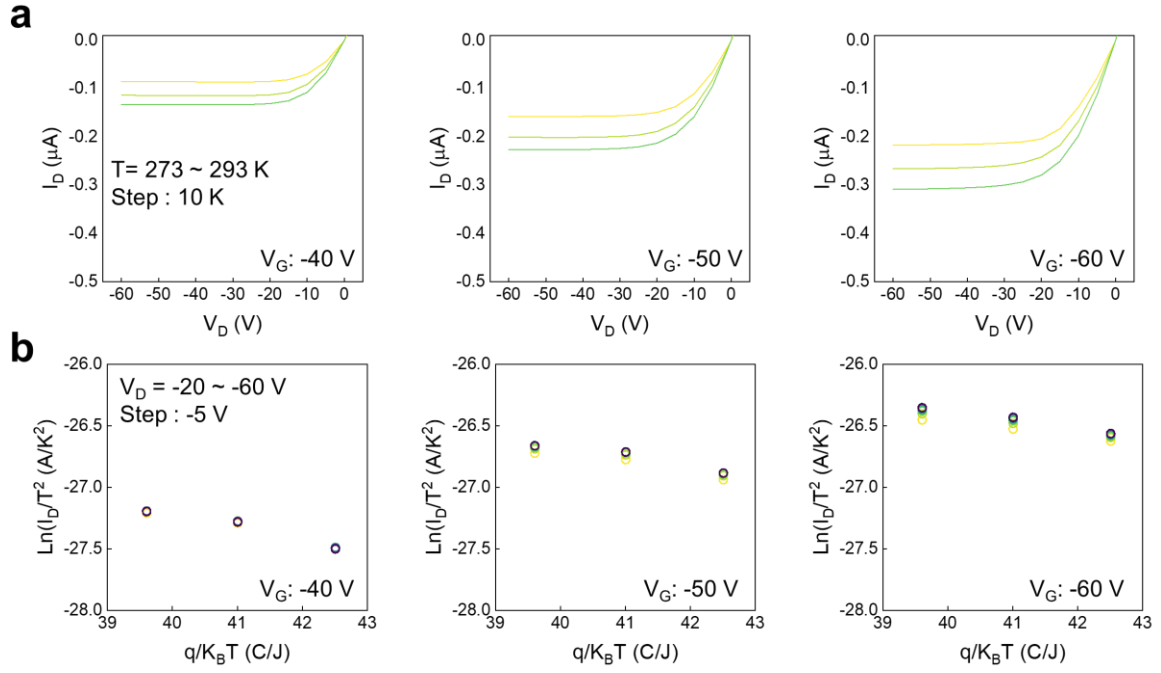
Supplementary Fig. 20. Optical images of about 5 μm damaged area with time flows during Chloroform solvent vapor treatment at room temperature after cutting: **a**, bright field images **b**, dark field images. Scale bar: 50 μm .



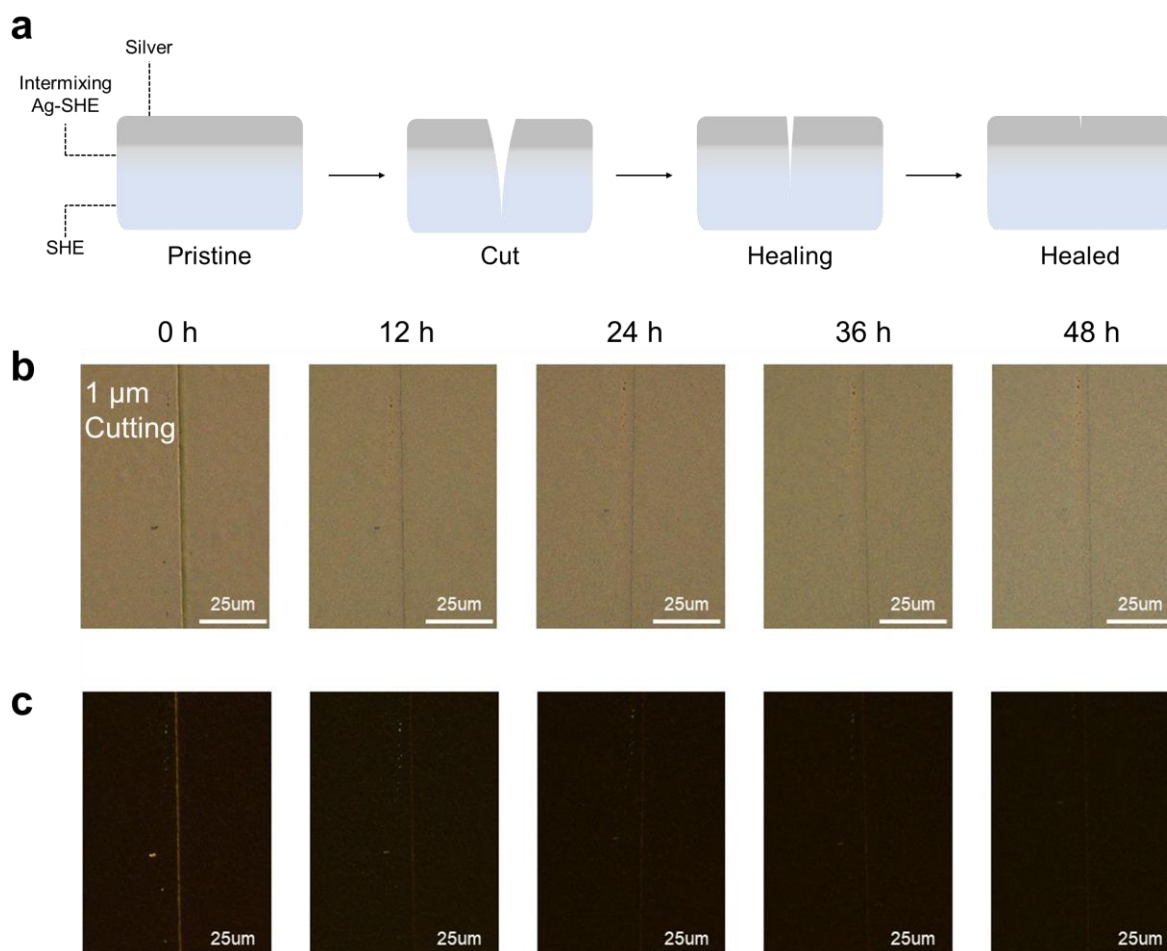
Supplementary Fig. 21. Optical images of about 9 μm damaged area with time flows during Chloroform solvent vapor treatment at room temperature after cutting: **a**, bright field images **b**, dark field images. Scale bar: 50 μm .



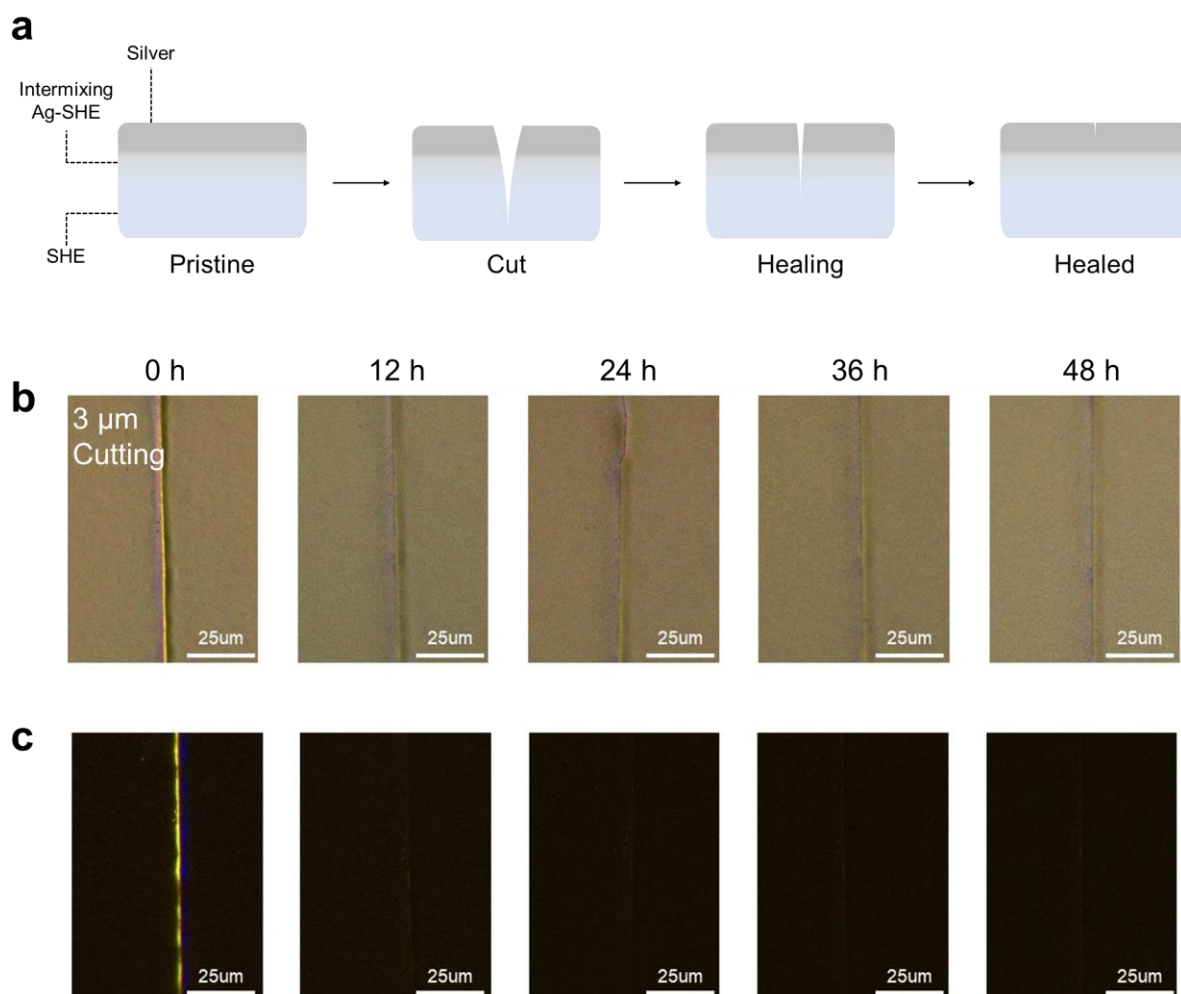
Supplementary Fig. 22. Schematic illustration of cutting process. a, Perpendicular and b, parallel cutting direction to channel direction. The transfer characteristics (V_D : -60 V) of c, perpendicular and d, parallel cutting direction to the channel direction with 1 μm cutting width.



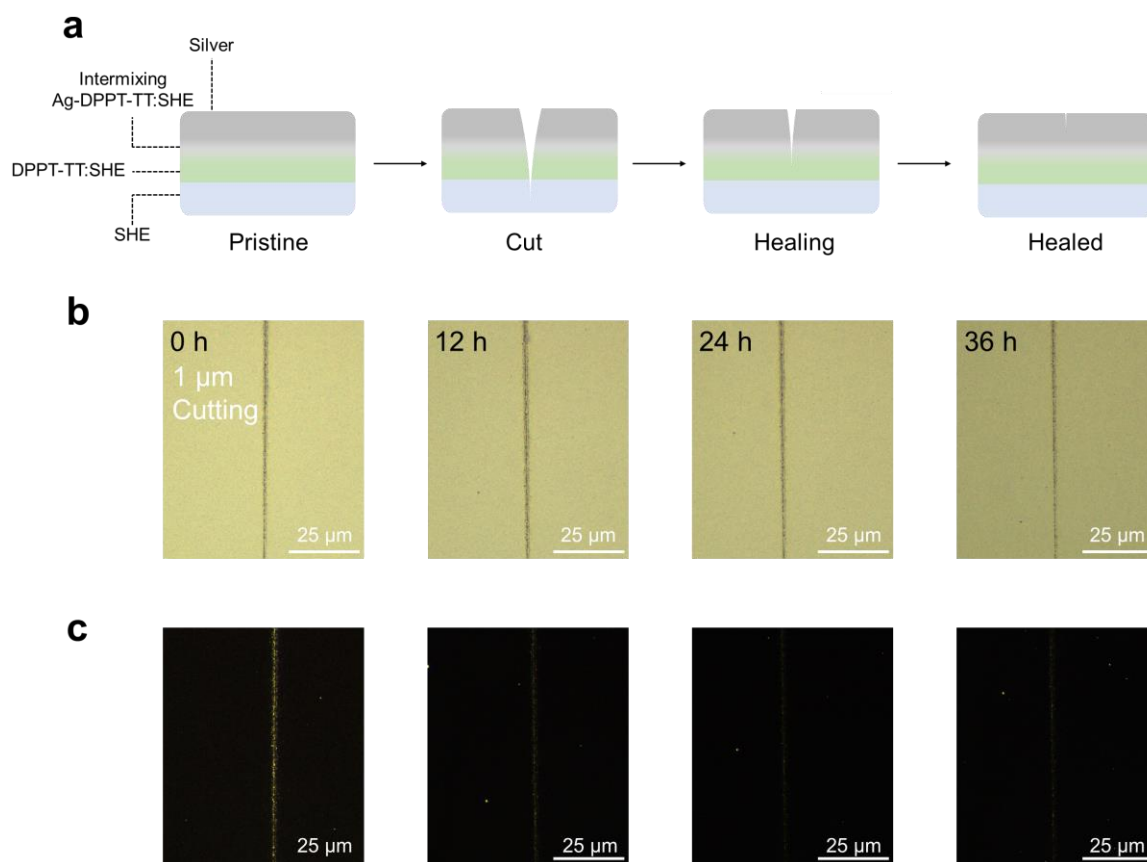
Supplementary Fig. 23. a, Output characteristics as a function of temperature with different gate voltage. **b**, Arrhenius fitting the $\ln(I_D/T^2)$ versus $q/K_B T$, where q and K_B are electrical constant and Boltzmann constant, respectively. Calculated effective Schottky barrier height against the drain voltage using below equation: $I_D = AA^*T^2 \exp(-q\Phi_B/k_B T)$, where A and A^* are barrier area and Richardson constant, respectively.



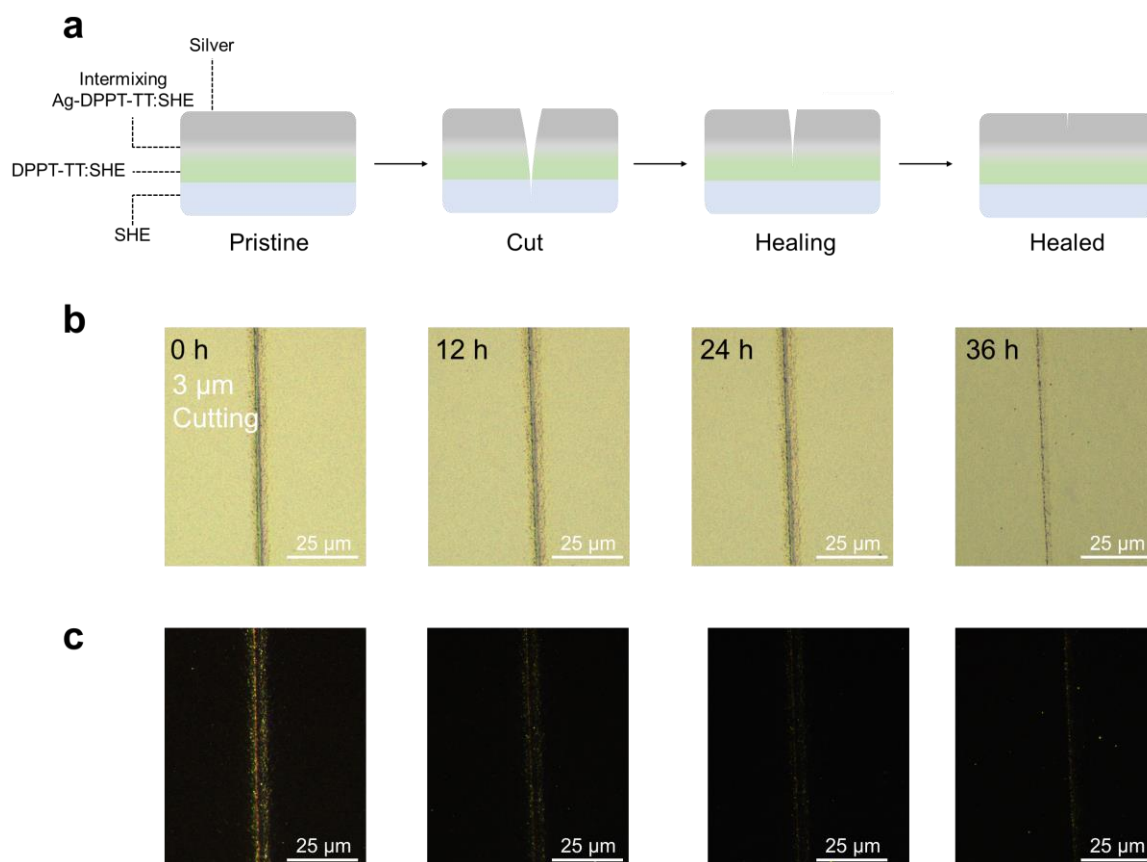
Supplementary Fig. 24. **a**, Schematic illustration of silver electrode healing on SHE substrate. The deposited-silver on SHE substrate was cut by surgical blade. After cutting, the silver was observed using optical microscope without any treatment. Optical images of silver electrode healing on SHE substrate at room temperature with time flows: **b**, Bright field images and **c**, dark field images. Cutting width is about 1 μm .



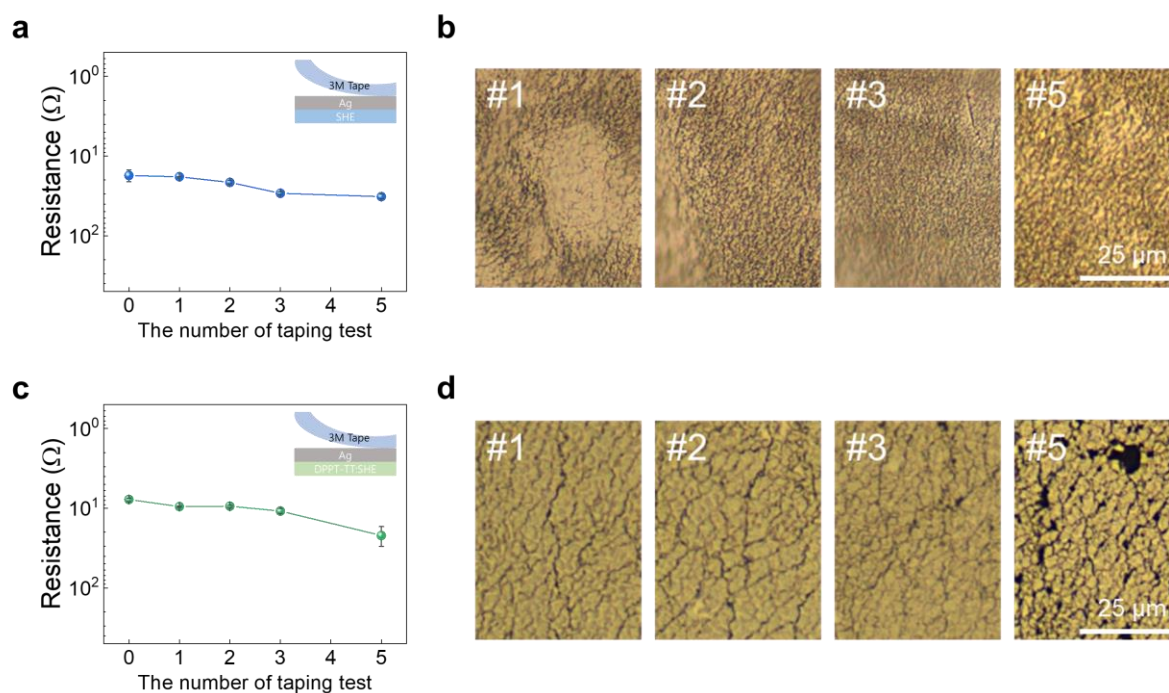
Supplementary Fig. 25. **a**, Schematic illustration of silver electrode healing on SHE substrate. Optical images of silver electrode healing on SHE substrate at room temperature with time flows: **b**, bright field images and **c**, dark field images. Cutting width is about 3 μm .



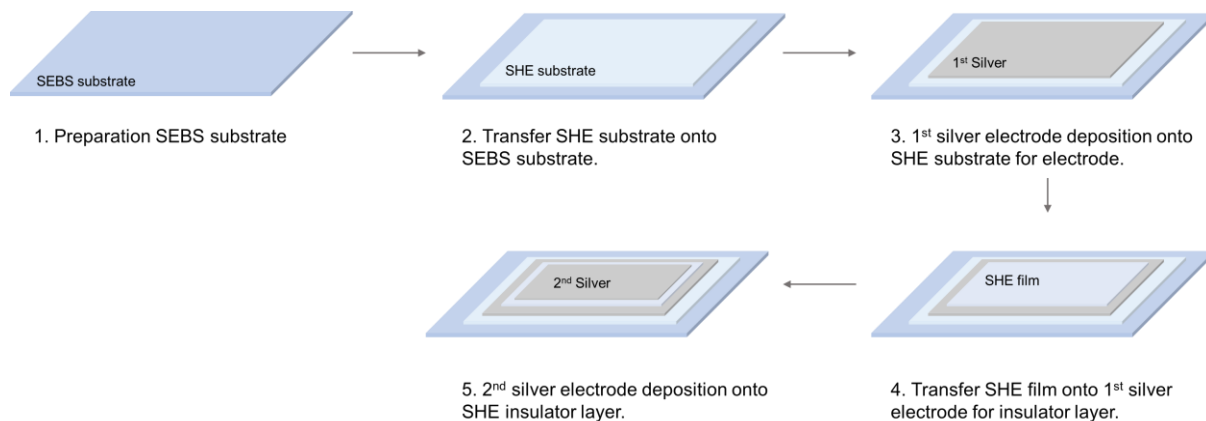
Supplementary Fig. 26. **a**, Schematic illustration of cut and autonomous-healing process of silver on DPPT-TT:SHE semiconducting film for source/drain electrode. The deposited silver was cut using surgical blade. Optical images of silver electrode healing on semiconductor at room temperature with time flows: **b**, bright field images and **c**, dark field images. Cutting width is about 1 μm .



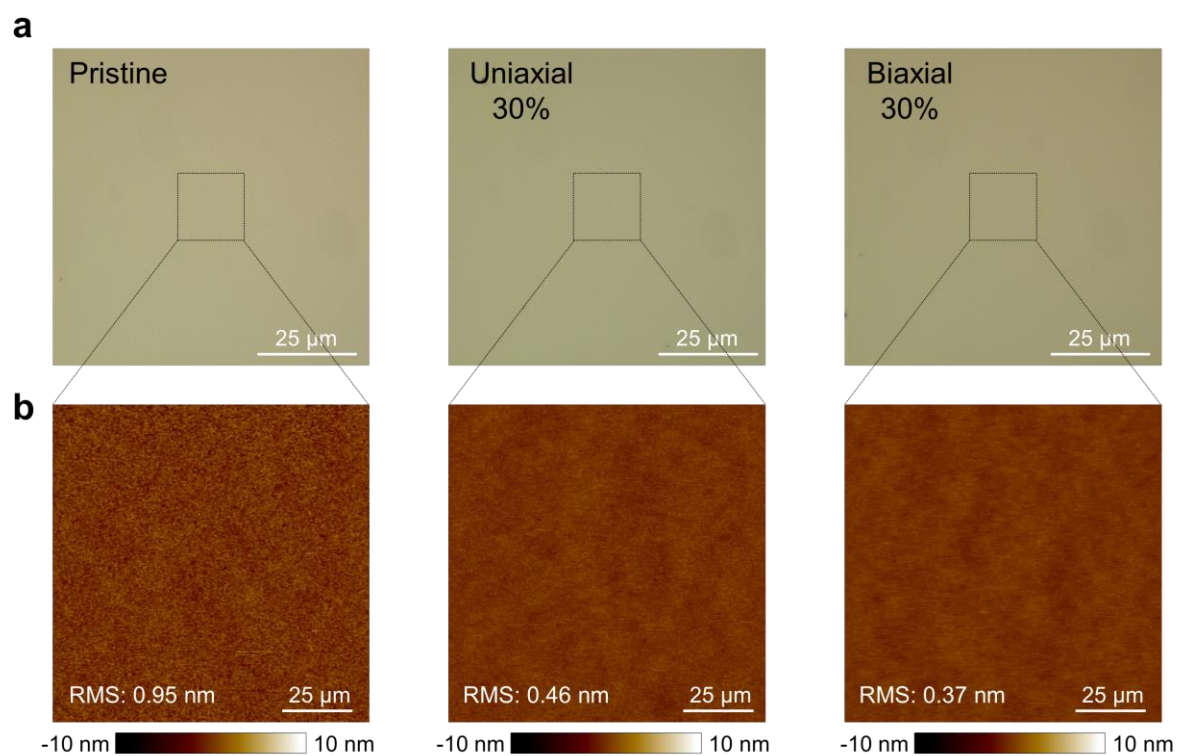
Supplementary Fig. 27. **a**, Schematic illustration of cut and autonomous-healing process of silver on DPPT-TT:SHE semiconducting film for source/drain electrode. The deposited silver was cut using surgical blade. Optical images of silver electrode healing on semiconductor at room temperature with time flows: **b**, bright field images and **c**, dark field images. Cutting width is about 3 μm .



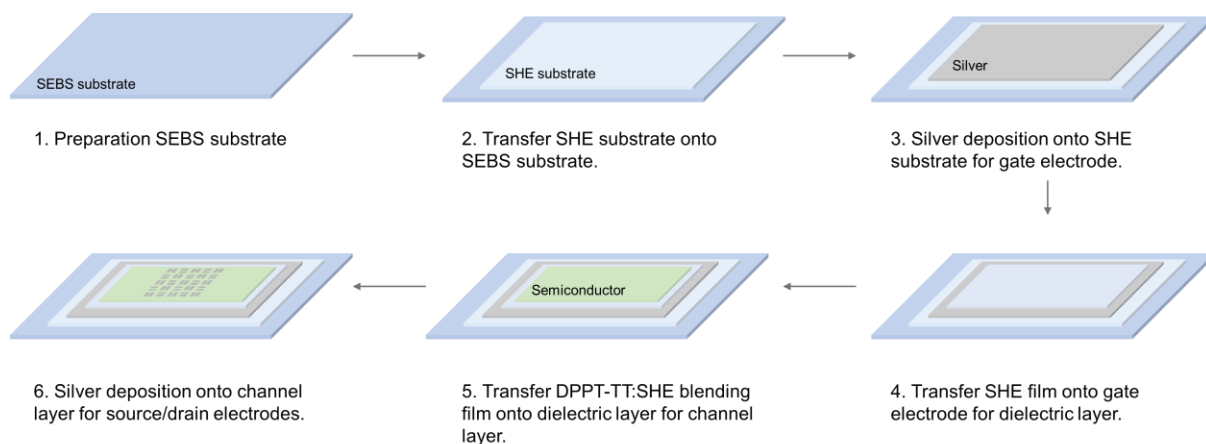
Supplementary Fig. 28. Resistance change of silver electrode (80 nm thick) on **a**, SHE and **c**, DPPT-TT:SHE film as a function of the number of taping test. The inset illustration shows Ag electrode delamination test using 3MTM tape. OM images of silver electrode on **b**, SHE and **d**, DPPT-TT:SHE film with the number of taping test. The electrical conductivity is still remained, even morphology is changed.



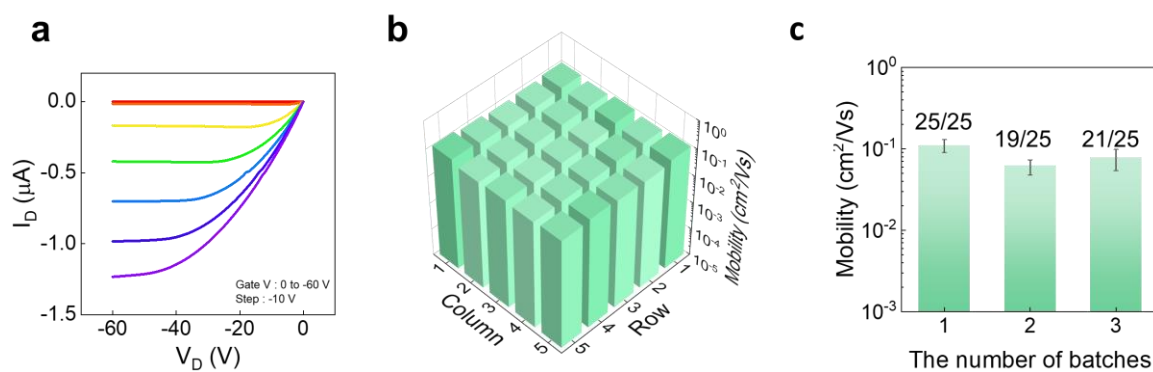
Supplementary Fig. 29. Fabrication steps for metal-insulator-metal (MIM) structure. The SHE substrate (1.0 μm) on OTS-treated SiO_2/Si wafer was directly transferred onto SEBS substrate. After transfer, the 1st silver (80 nm) was deposited on to SHE substrate. The another SHE film (1.5 μm) on OTS-treated SiO_2/Si wafer was directly transferred onto 1st silver electrode. The 2nd silver electrode (80 nm) was deposited onto SHE films.



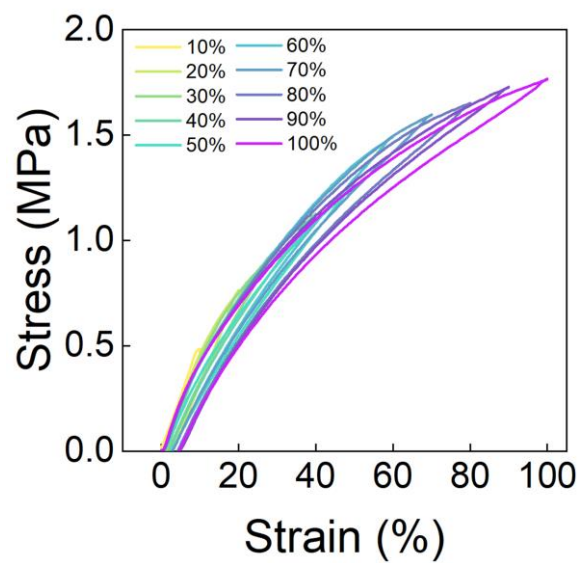
Supplementary Fig. 30. a, OM and **b**, AFM height images of SHE dielectric films on 0%, 30% uniaxial and 30% biaxially strain.



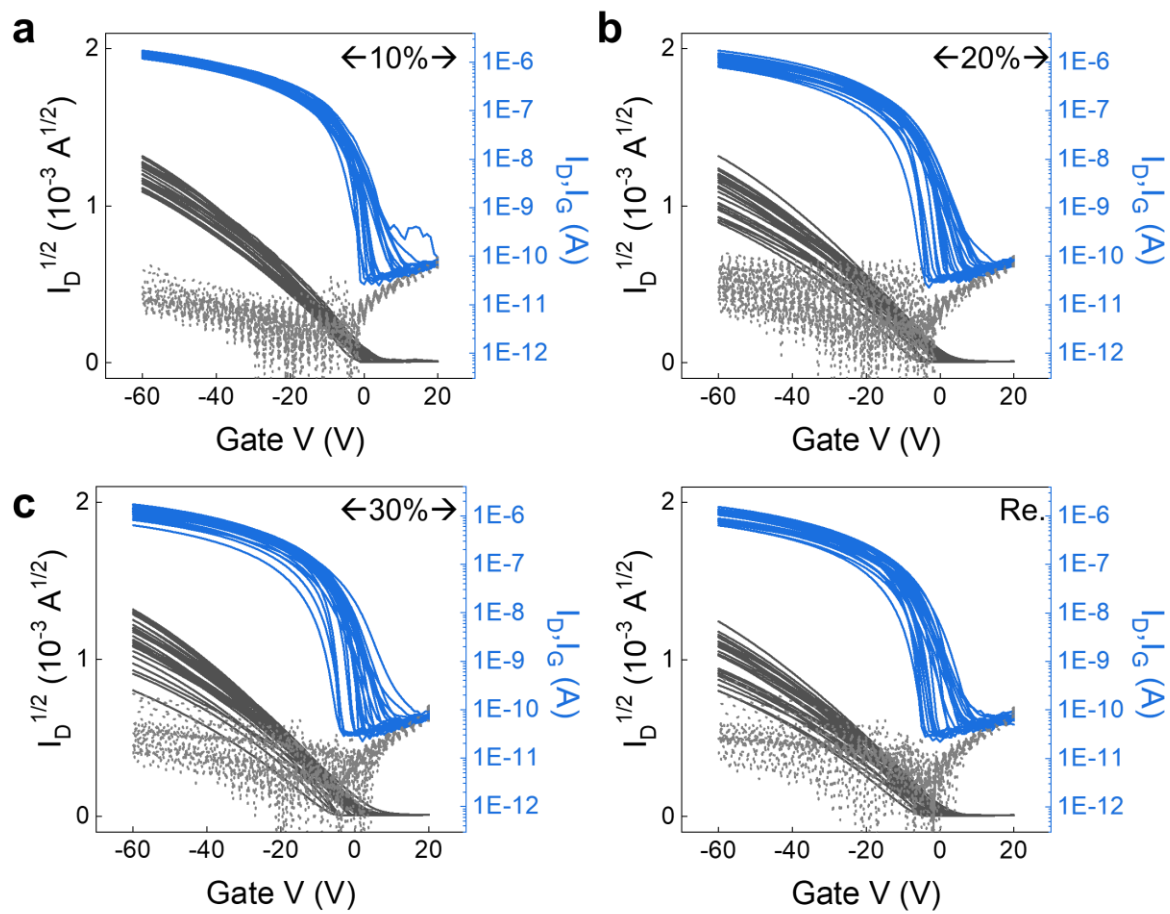
Supplementary Fig. 31. Fabrication steps for stretchable and self-healable passive arrays. The silver gate (80 nm) was deposited onto SHE substrate (1.0 μm) with thermal deposition. After fabrication of gate, SHE dielectric (1.5 μm) and semiconductor (100 nm) on OTS-treated SiO_2/Si wafer was directly transferred onto gate electrode. Finally, silver S/D electrodes (80 nm) were deposited onto semiconductor.



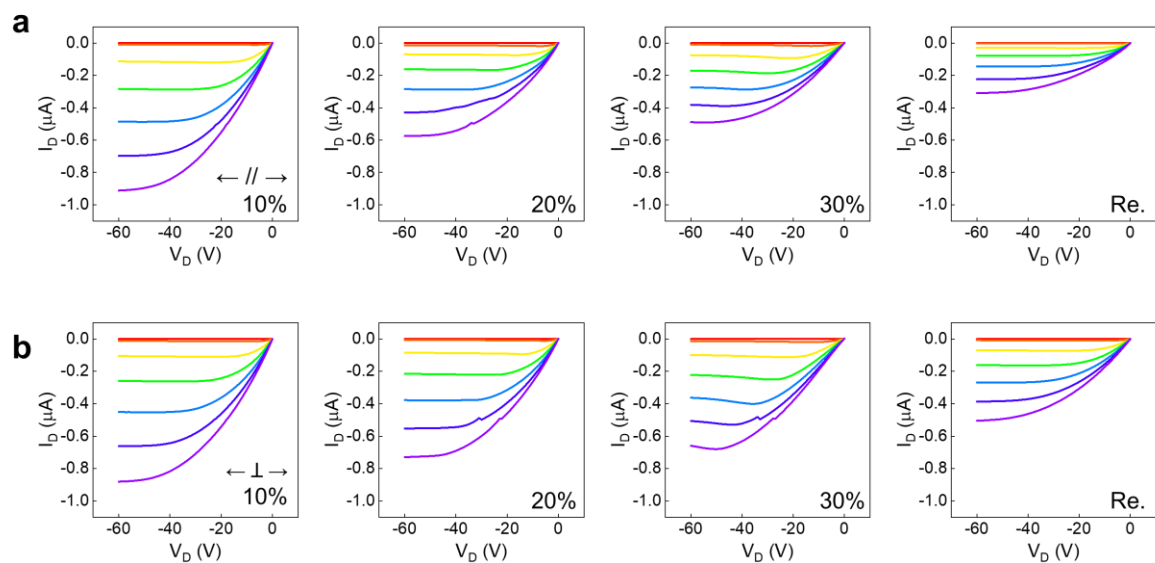
Supplementary Fig. 32. **a**, Output characteristic and **b**, field effect saturation mobility mapping of fully stretchable and self-healable passive arrays. **c**, Mobility deviation in three different batches.



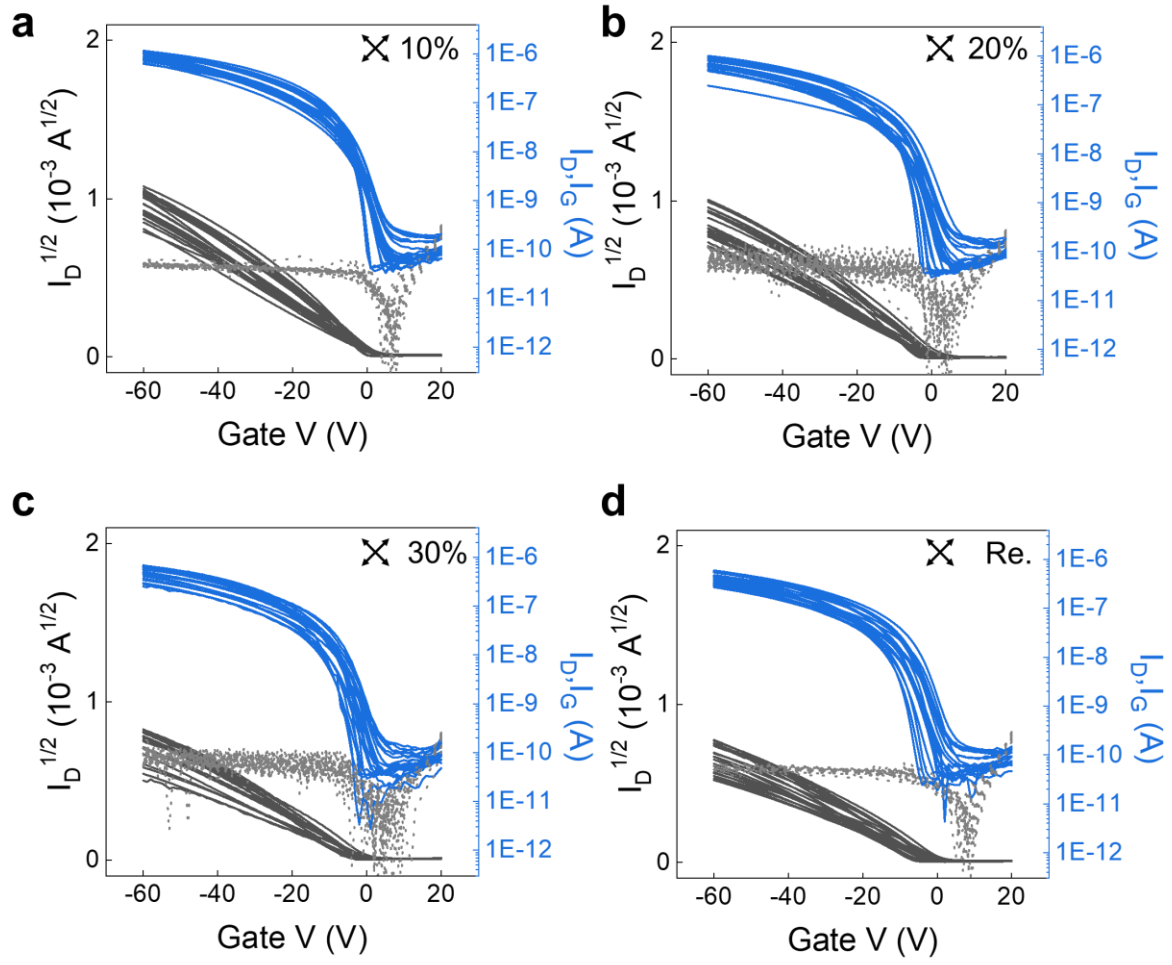
Supplementary Fig. 33. Cyclic strain-stress curves from 10% to 100% strain of fully stretchable and self-healable passive transistor array (5×5) module.



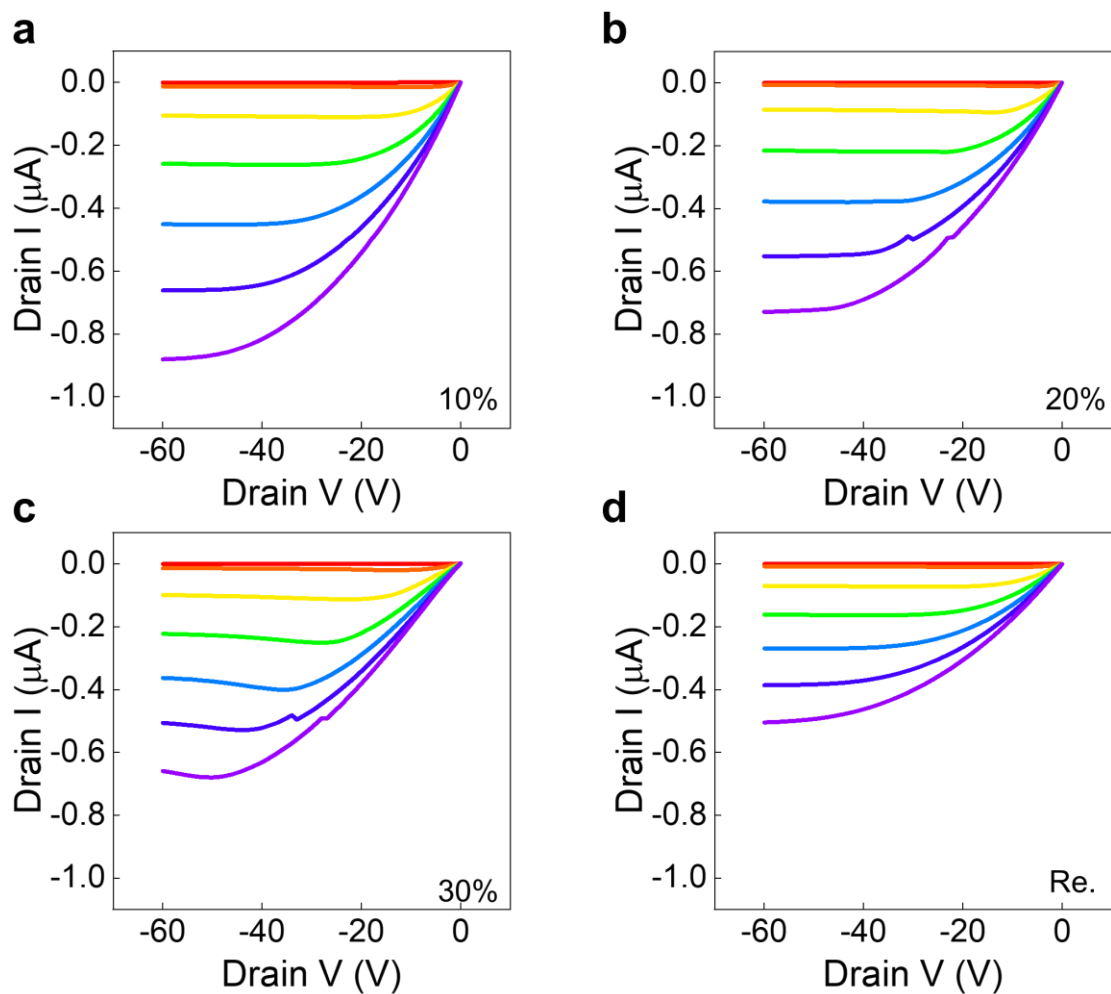
Supplementary Fig. 34. Transfer characteristics of fully stretchable and self-healable passive transistor array (5×5) under **a**, 10%, **b**, 20%, **c**, 30% and **d**, 30%-release of uniaxial strain.



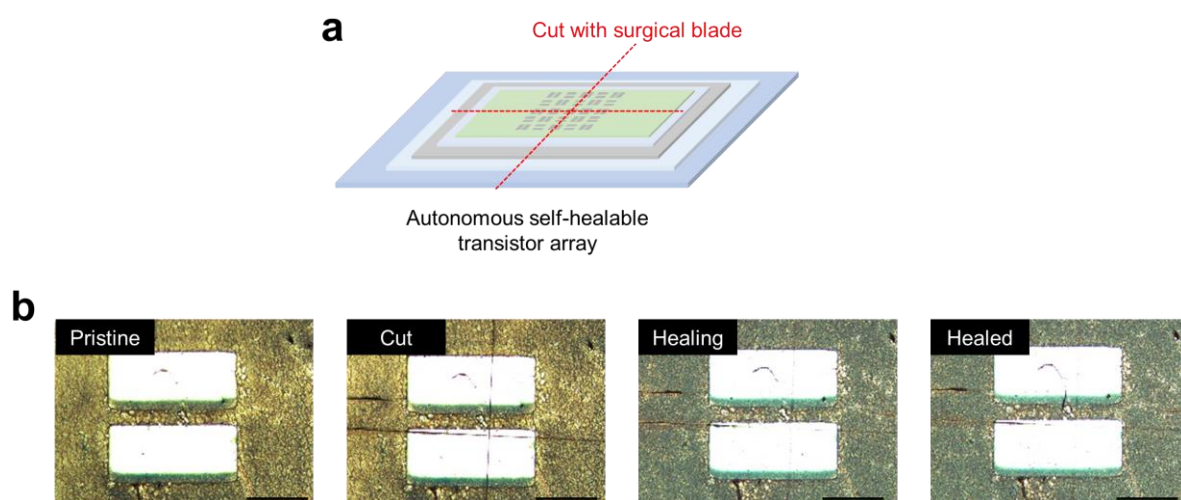
Supplementary Fig. 35. Representative output characteristics (V_G : 0 to -60 V, step: -10 V) of fully stretchable and self-healable passive arrays under uniaxial strain with **a**, parallel and **b**, perpendicular direction to channel direction.



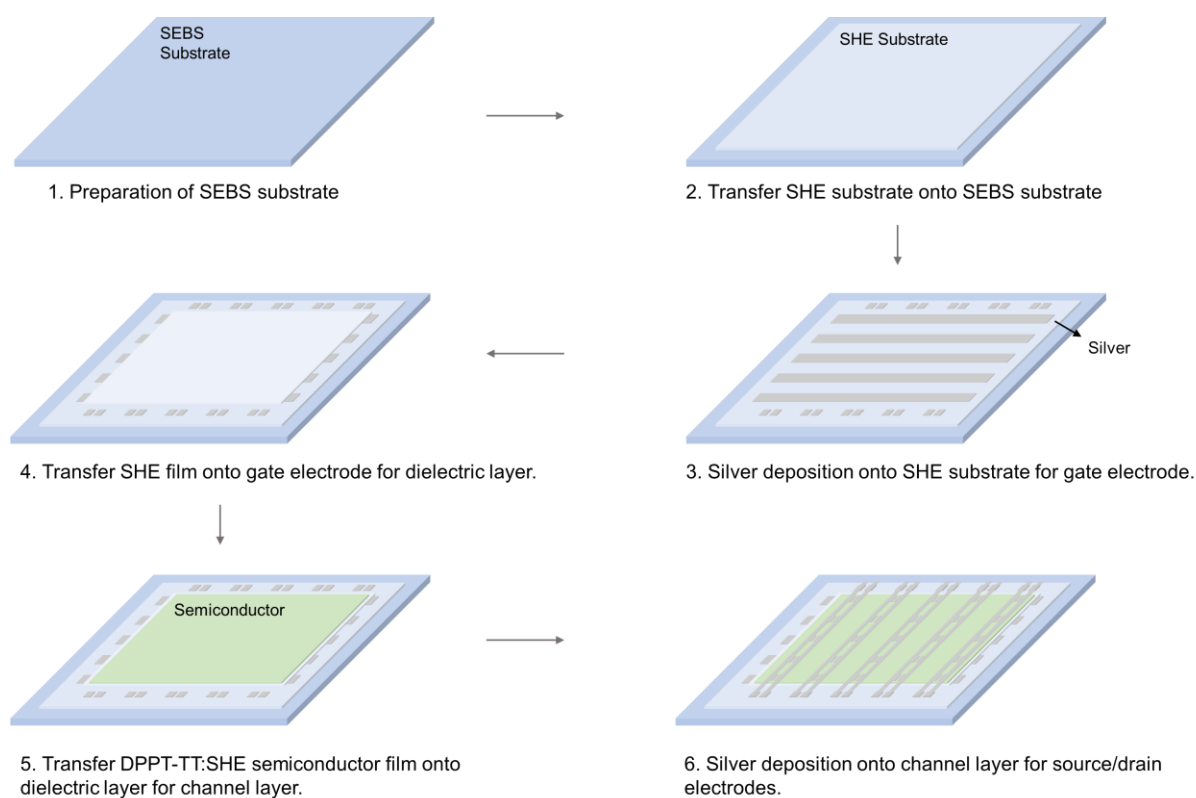
Supplementary Fig. 36. Transfer characteristics of fully stretchable and self-healable passive transistor array (5×5) under **a**, 10%, **b**, 20%, **c**, 30% and **d**, 30%-release of biaxial strain.



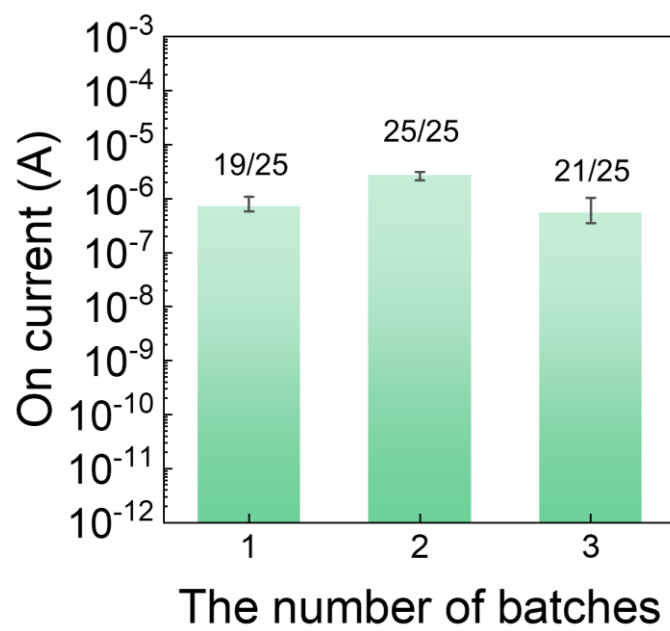
Supplementary Fig. 37. Representative output characteristics of fully stretchable and self-healable passive arrays under **a**, 10%, **b**, 20%, **c**, 30%, and **d**, 30%-release of biaxial strain.



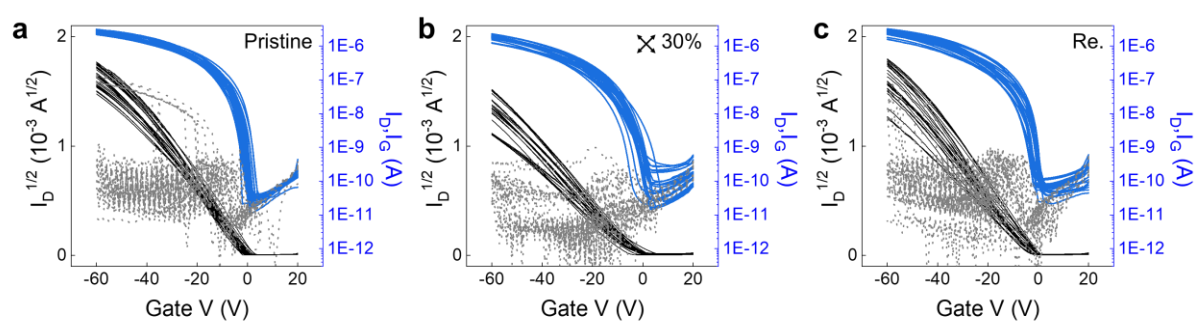
Supplementary Fig. 38. a, Schematic illustration for cross-bar shape cutting on transistor array. **b,** OM images of a unit device in center of transistor array: pristine, cut, healing (24 h) and healed (48 h) at room temperature. The scale bar is 500 μm .



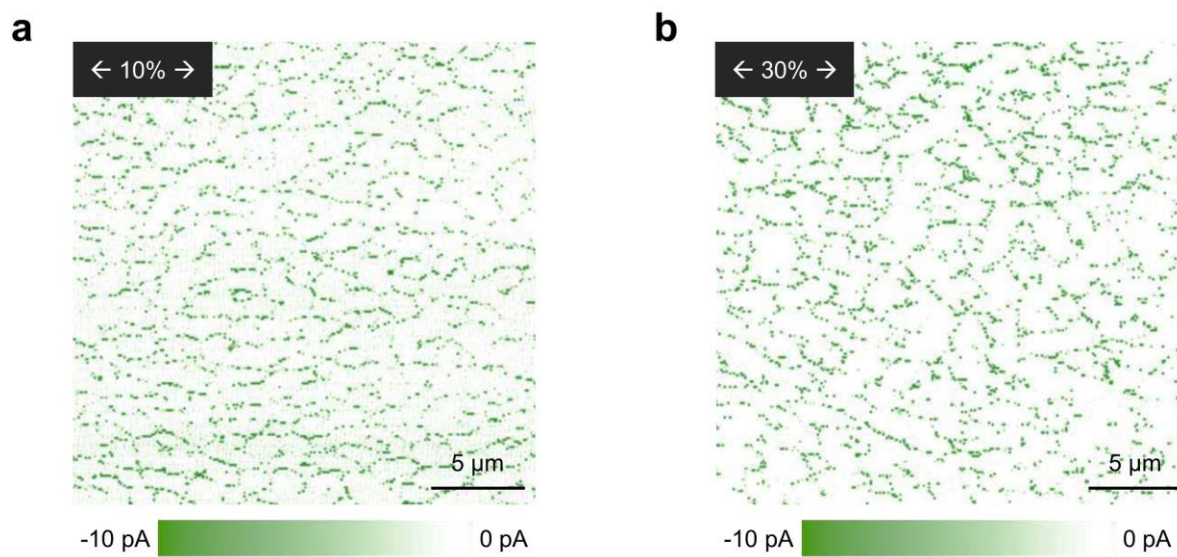
Supplementary Fig. 39. Fabrication steps for stretchable and self-healable active matrix arrays. Silver gate electrode (80 nm) was deposited onto SHE substrate (1.0 μm). SHE dielectric (2.1 μm) and semiconducting film (100 nm) on OTS-treated SiO_2/Si wafer was transferred directly onto silver electrode. Finally, silver S/D electrodes (80 nm) were deposited.



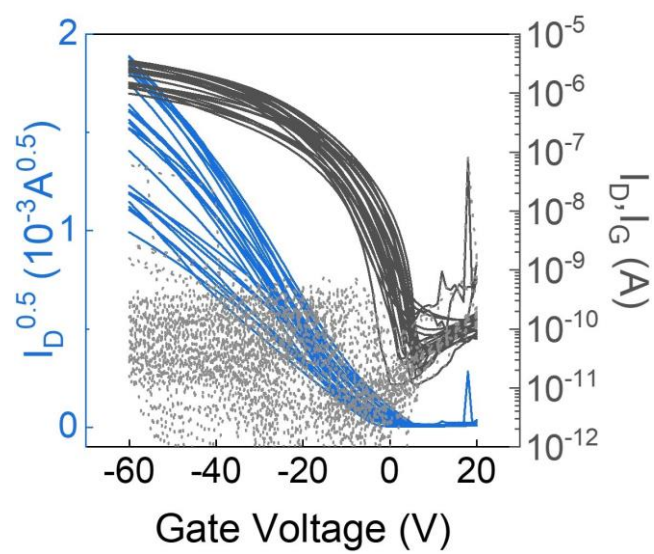
Supplementary Fig. 40. Average on-current ($V_D, V_G = -60$ V) of stretchable and self-healable active-matrix arrays in three different batches. (x/y, x: working devices, and y: total devices)



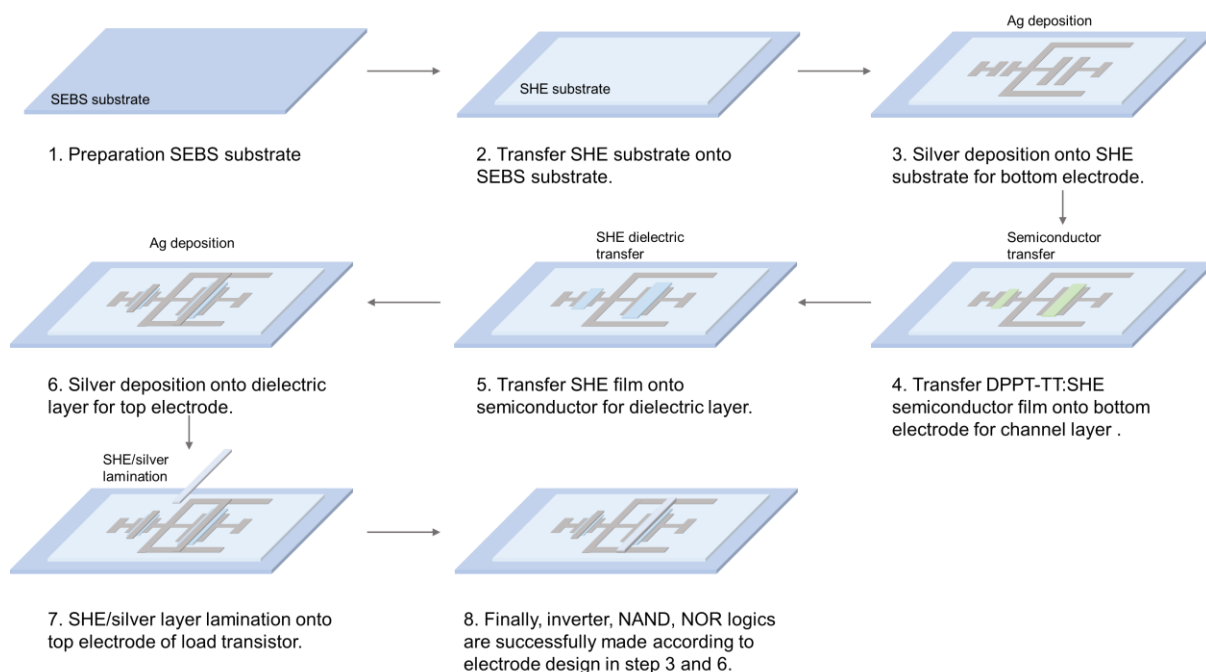
Supplementary Fig. 41. Transfer characteristics for twenty-five unit devices of stretchable and self-healable active matrix arrays under **a**, pristine, **b**, 30% biaxially stretched, **c**, 30%-released states.



Supplementary Fig. 42. Conductive-AFM images of semiconducting films upon **a**, 10%, and **b**, 30% uniaxial strain.

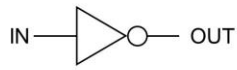


Supplementary Fig. 43. Transfer characteristics of twenty-five unit devices of active matrix arrays after healing at room temperature (48 h).



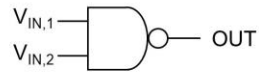
Supplementary Fig. 44. Fabrication steps for stretchable and self-healable inverter, NAND, NOR logics. Silver S/D electrode (80 nm) was deposited onto SHE substrate (1.0 μm). After deposition, semiconductor (100 nm) and SHE dielectric (2.1 μm) were directly transferred onto S/D electrode. Silver gate (80 nm) was deposited onto dielectric film, and SHE/silver film (1.0 μm /80 nm) was laminated onto gate electrode of load transistor.

a Inverter



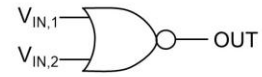
V_{IN}	V_{OUT}
1	0
0	1

b NAND



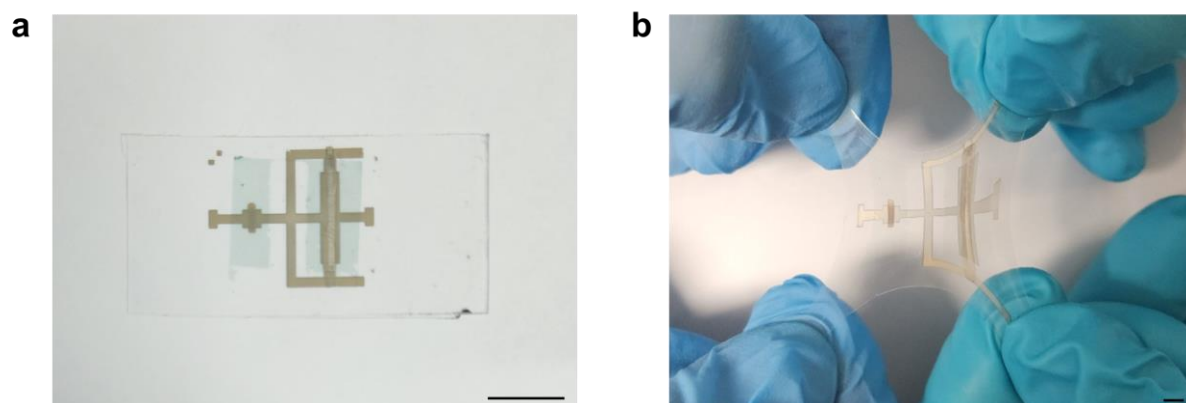
$V_{IN,1}$	$V_{IN,2}$	V_{OUT}
0	0	1
0	1	1
1	0	1
1	1	0

c NOR

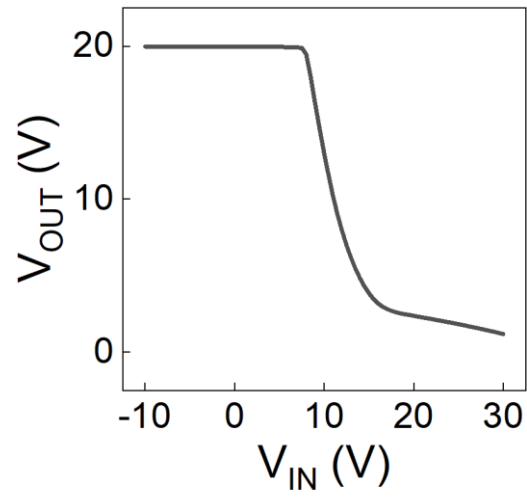


$V_{IN,1}$	$V_{IN,2}$	V_{OUT}
0	0	1
0	1	0
1	0	0
1	1	0

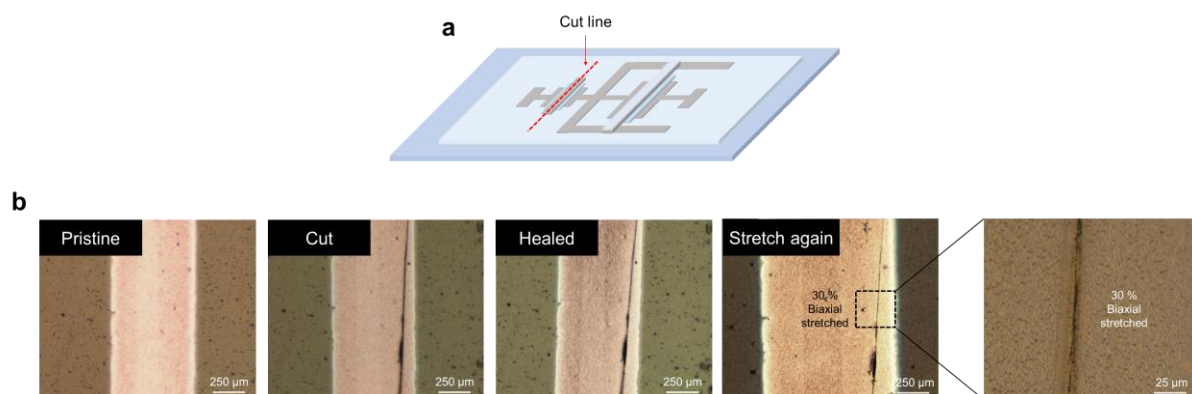
Supplementary Fig. 45. Circuit diagrams of **a**, inverter, **b**, NAND and **c**, NOR logic devices.



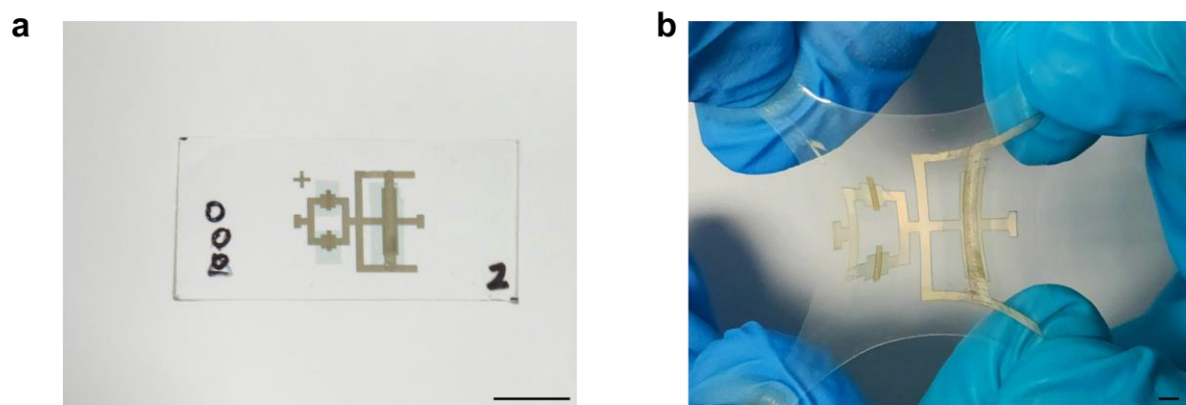
Supplementary Fig. 46. Photographs of **a**, pristine and **b**, biaxially stretched inverter devices. The scale bar is 1 cm.



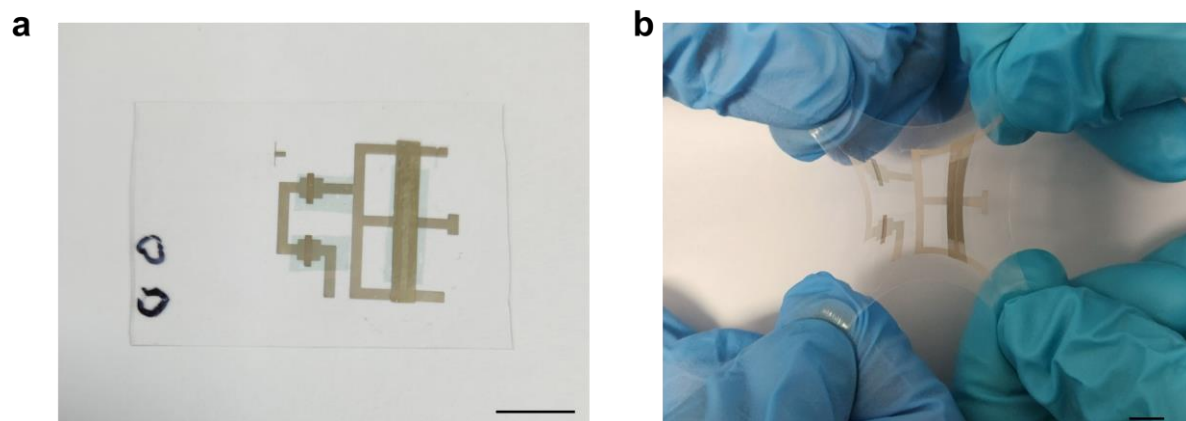
Supplementary Fig. 47. Transfer characteristic of inverter device under 10% biaxial strain at $V_{DD} = 20$ V.



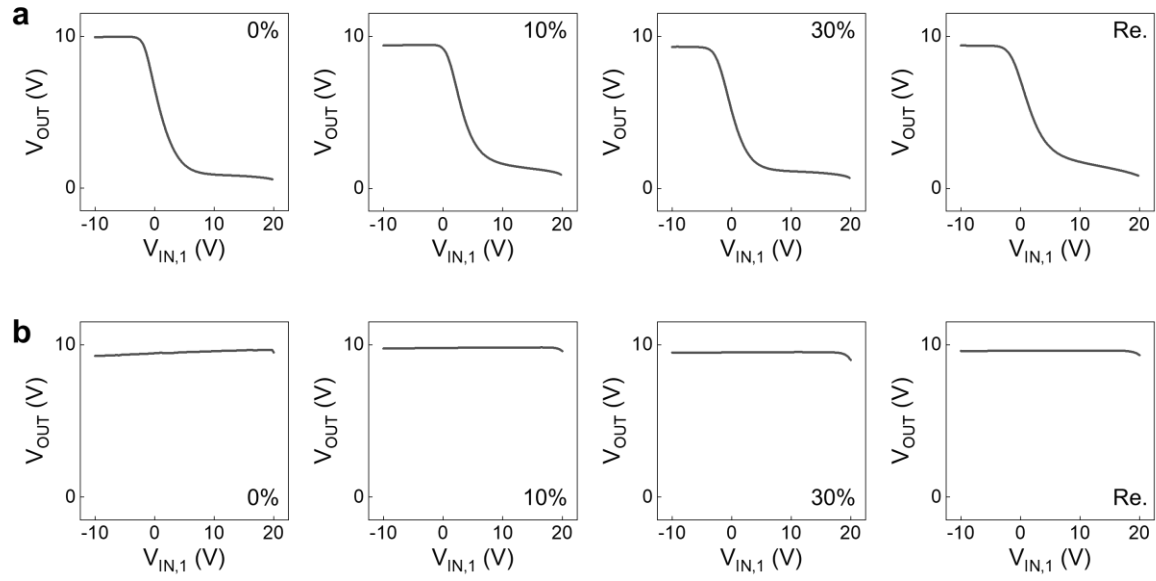
Supplementary Fig. 48. a, Schematic illustration of cutting process for inverter device. Drive transistor of device was cut using surgical blade. **b**, OM images of drive transistor under pristine, cut, healed (after 48 h) and stretched again after healed states.



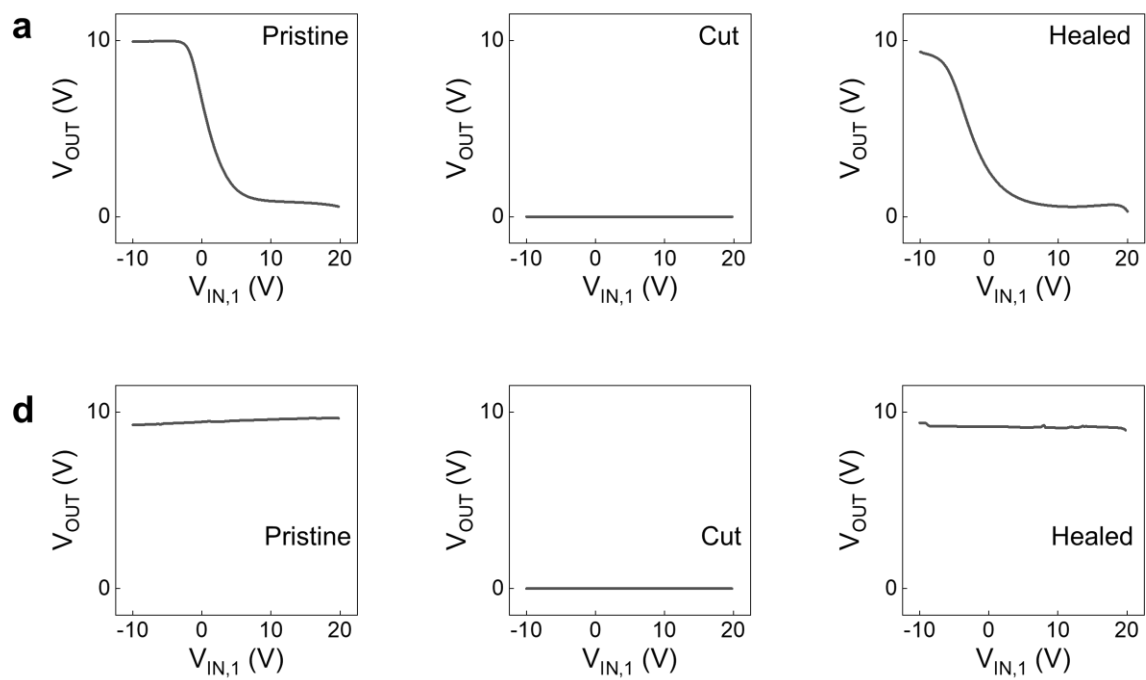
Supplementary Fig. 49. Photographs of **a**, pristine and **b**, biaxially stretched NAND devices. The scale bar is 1 cm.



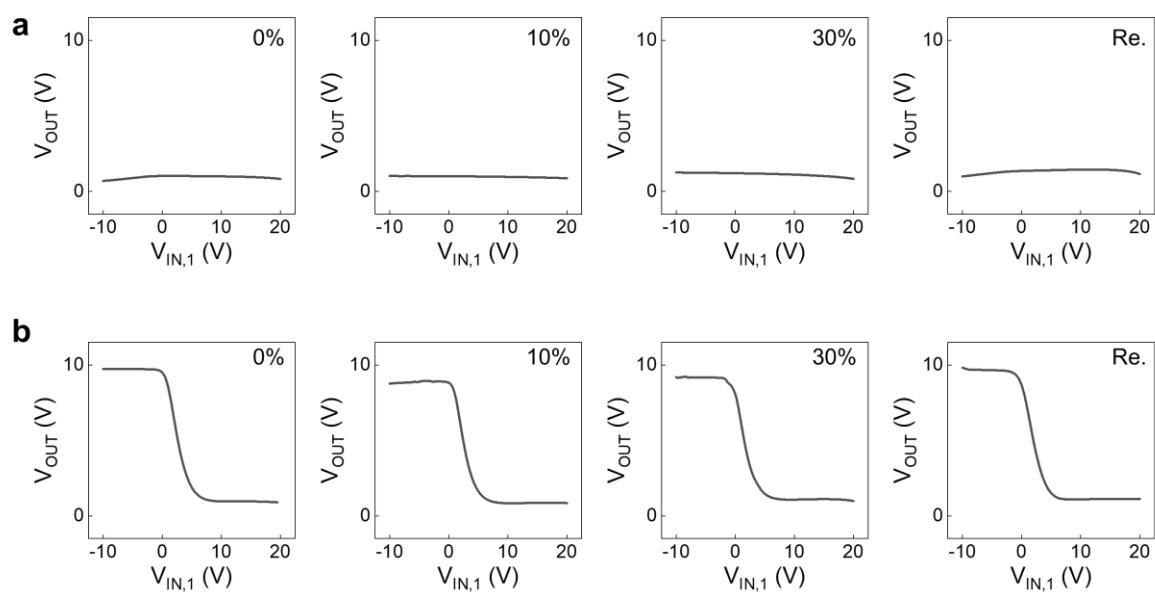
Supplementary Fig. 50. Photographs of **a**, pristine and **b**, biaxially stretched NOR devices. The scale bar is 1 cm.



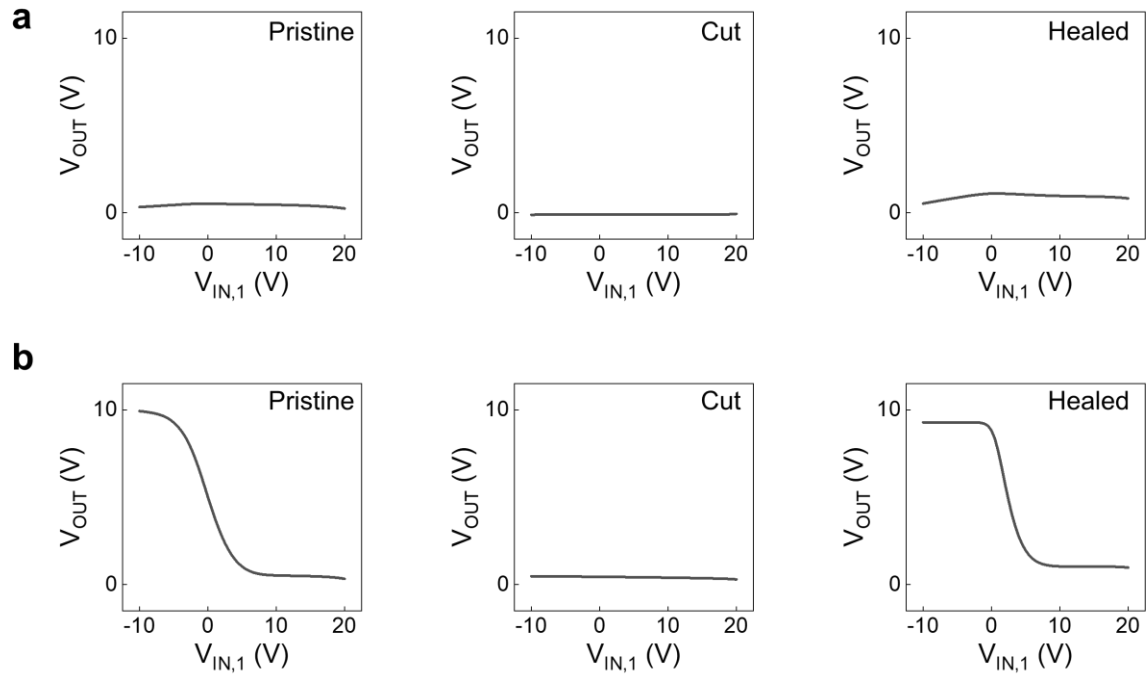
Supplementary Fig. 51. VTCs of NAND devices with various biaxial strain. Output voltages under 0 % to 30 % strain and released at **a**, $V_{IN,2} = 20$ V and **b**, $V_{IN,2} = -10$ V.



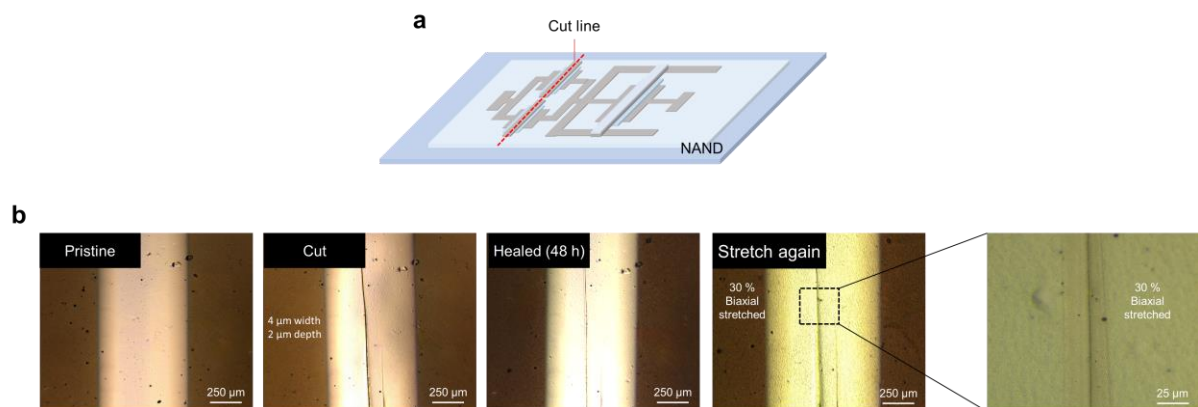
Supplementary Fig. 52. VTCs of NAND devices under autonomous self-healing process. Output voltages of pristine, cut, healed (48 h) states at **a**, $V_{IN,2} = 20$ V and **b**, $V_{IN,2} = -10$ V.



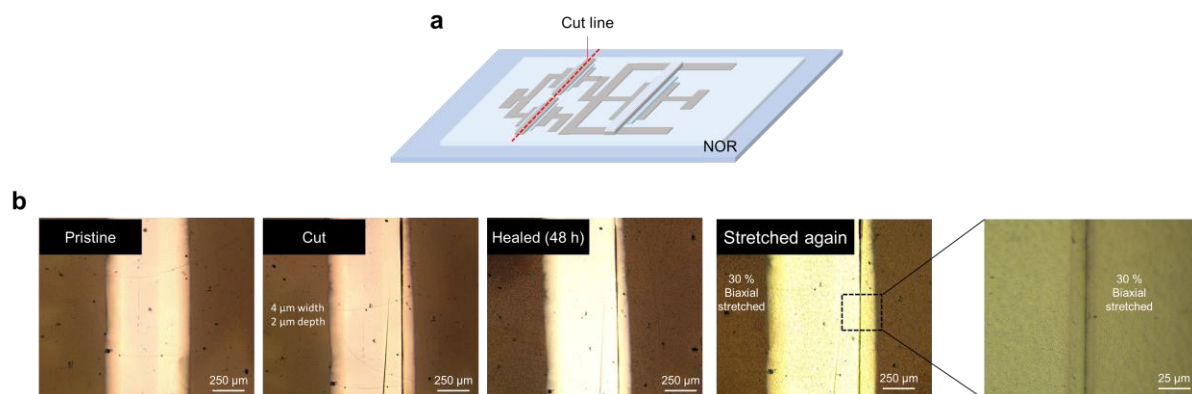
Supplementary Fig. 53. VTCs of NOR devices with various biaxial strain. Output voltages under 0 % to 30 % strain and released states at **a**, $V_{IN,2} = 20$ V and **b**, $V_{IN,2} = -10$ V.



Supplementary Fig. 54. VTCs of NOR devices under autonomous self-healing process. Output voltages of pristine, cut, healed (48 h) states at **a**, $V_{IN,2} = 20$ V and **b**, $V_{IN,2} = -10$ V.



Supplementary Fig. 55. a, Schematic illustration of cutting process for NAND device. Both drive transistors of NAND were cut using surgical blade. **b**, OM images of drive transistor under pristine, cut, healed (after 48 h) states.



Supplementary Fig. 56. a, Schematic illustration of cutting process for NOR device. Both drive transistors of NOR were cut using surgical blade. **b**, OM images of drive transistor under pristine, cut, healed (after 48 h) and stretched again after healed states.

Supplementary Table 1. Device geometry and dielectric capacitance under strain.

Stretching	Strain (%)	Channel length (μm)	Channel width (μm)	Capacitance (nF/cm ²)
Uniaxial ()	0	150	1000	1.75
	10	176	918	1.80
	20	193	883	1.88
	30	202	857	1.9
Uniaxial (=)	0	150	1000	1.75
	10	137	1180	1.85
	20	132	1180	1.85
	30	130	1257	1.93
Biaxial (×)	0	150	1000	1.75
	10	176	1032	1.86
	20	188	1092	1.94
	30	199	1148	2.24

Note that the capacitance values of gate dielectric were obtained at 1 kHz.

Supplementary Table 2. Comparison of our performance and previously reported works.

Structure	Mobility (cm ² /Vs)	Healing components	Healing method	Healing scale	Healing time	Stretchability	Ref.
	0.11	All components (Source/drain, semiconductor, dielectric, gate, and substrate)	Autonomous healing	5 μm	36 hour	Uniaxial: 30% Biaxial: 30%	This work
	0.286	Semiconductor	Solvent vapor and heat treatments	0.05 μm	N/A	Uniaxial: 100% Biaxial: N/A	#4
	0.076	Semiconductor	Autonomous healing	0.2 μm	24 hour	Uniaxial: 100% Biaxial: N/A	#5
	0.002	Semiconductor	Solvent vapor and heat treatments	0.05 μm	N/A	Uniaxial: 20% Biaxial: N/A	#6
	0.16	Semiconductor	Autonomous healing	N/A	Few seconds	Only semiconductor Uniaxial: 150% Biaxial: N/A	#7
	0.4	Semiconductor	Melt process	20 μm	N/A	Non-stretchable	#8

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