Electronic Supplementary Information

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Experimental Section:

Preparation of the NSSU and LSSU units: A mixture of uncured Ecoflex® silicone (all Ecoflex® used in this work contains Part A and Part B with 1:1 mass ratio, Smooth-On, Inc.) and red soap dye (containing titanium dioxide, mica, iron oxide, and tin oxide, MyoMakeup.com) with a mass ratio of 10:1 was drop cast on a VHB™ 4905 double-sided adhesive tape (thickness ≈ 0.5 mm, 3M) with a thickness of ca. 0.1 mm, followed by curing at 80 °C for 2 hours. A layer of mirror chrome (containing metal flakes, thickness ≈ 40 nm, Spaz Stix, Inc.) was then spray coated onto the other side of the VHB™ 4905 double-sided adhesive tape by an airbrush style spray-gun (Master Airbrush G444-SET, equipped with a 0.5 mm needle nozzle and a Royal Mini Air Compressor, TC-20B, 10 mg/mL TiO_2/PVB ethanol suspension), followed by the spray coating of TiO_2 (99.9%, CR828, Tronox) and polyvinyl butyral (PVB) (Mowital® B 60 HH, Kuraray, mass ratio of PVB/TiO_2= 4:1) with a thickness of ca. 890 nm to form a light shield layer. A layer of pure Ecoflex® (thickness ≈ 6.6 μm, 100 mg/mL of Ecoflex®/hexane solution) was then spray coated atop the PVB/TiO_2 layer and cured at 80 °C for 30 min followed by a 20 min UVO treatment in a Novascan PSD digital UV ozone system. Subsequently, a homogeneous thermochromic and photochromic pigment mixture layer (thickness ≈ 3.3 μm) with a mass ratio of 1:1 was spray coated atop the cured and UVO treated Ecoflex® layer (the thermochromic pigment was purchased from Karlsson Robotics and the photochromic pigment was purchased from Glomania; both the thermochromic and photochromic pigments used in this work contain a mixture of Leuco dyes, weak acids and salts microencapsulated in polymer; 10 g/mL pigment ethanol suspension). A layer of uncured Ecoflex® with a thickness of ca. 0.39 mm was cast atop the pigment mixture layer and placed in a refrigerator set at -10 °C for 4 hours to allow the uncured Ecoflex® to penetrate into the porous spacing between the pigment particles followed by curing at 80 °C for 2 h. The preparation of the LSSU is the same as the aforementioned procedures except for subjected to a 150% pre-stretching strain prior to spray coating the light shielding layer and released to 0% strain prior to final addition of the thick Ecoflex® layer.

Preparation of the LED set with electrochromic characteristic and stretchable conductor for the interactive electronics: An ITO coated PET film (Sigma Aldrich, surface resistivity ≈ 60 Ω/sq, ITO coating thickness ≈ 130 nm, PET film thickness ≈ 0.127 mm) was attached to a glass slide (size: 3.0 cm×2.5 cm*)* via a double-sided adhesive tape (3M 9474LE double-sided adhesive). A layer of PEDOT: PSS (1.0 wt. % in H_2O, purchased from Sigma Aldrich, resistance ≈ 50 to 120 Ω/sq, thickness ≈ 300 nm, area ≈ 1 cm×1 cm) was cast and dried atop the ITO coated PET film on the position as indicated in Figure S16. A gap of ca. 0.5 mm was created across the thickness of the conductive PEDOT: PSS layer and the PET film to allow for the formation of two identical separated electrodes followed by casting a layer of PMMA/$LiClO_4$/propylene carbonate polymer gel electrolyte (PMMA, Plexiglas® V825, $M_w \approx 100{,}000$, $LiClO_4$ and propylene carbonate were purchased from Alfa Aesar, mass ratio of PMMA/$LiClO_4$/propylene carbonate = 3.8:1.0:11.6) as shown in Figure S16 and then encapsulated by PDMS (Sylgard® 184, base to curing agent = 10 to 1, cured at 55 °C for 8 h). A LilyPad LED lamp (purchased from Sparkfun) was connected with the two separated ITO electrodes via a conductive adhesive (purchased from Atom Adhesives, AA-DUCT 907 Silver Conductive Epoxy). To prepare the stretchable conductor, the VHB™ 4905 double-sided adhesive tape (size: 4.0 cm×2.5 cm) was pre-stretched to 450% strain and attached to a releasable rigid frame. The conductive thread spun from stainless steel fiber (purchased from Sparkfun, diameter: ca. 0.12 mm) was adhered to the stretched VHB™ tape and roll pressed by a glass rod. Another VHB™ 4905 double-sided adhesive tape was also pre-stretched to 450% and attached atop the conductive thread and roll pressed by a glass rod to make a sandwich-like structure followed by releasing to 0% strain.

Preparation of the interactive electronics: The NSSU, LSSU (both having a size of 5.5 cm×0.6 cm), an LED device with electrochromic unit (3.0 cm×2.5 cm), and a stretchable conductor (5.5 cm×2.5 cm) were attached to a layer of VHB™ 4905 double-sided adhesive tape (14 cm×2.5 cm) with a layout as shown in Figure 2h. The thread of the stretchable conductor was connected to the ITO coated PET by silver conductive epoxy and the edges of the stretchable conductor (VHB™ part) were adhered to the LED device with a silicone adhesive (Sil-Poxy, Smooth-On, Inc.).

Preparation of thermal mapping unit and the HSSU for the wearable motion/environmental monitoring (WMEM) device: To prepare the thermal mapping unit, a homogeneous thermochromic pigment mixture layer (thickness ≈ 3.3 μm) was spray coated atop the cured and UVO treated Ecoflex®/TiO_2 (thickness ≈ 0.5mm, mass ratio = 10:1) substrate. The thermochromic mixture layer contains three types of thermochromic pigments as shown in Figure S25 with transition temperatures of 15, 33, and 65 ºC, respectively, and the mass ratio of them was 4:4:1. (thermochromic pigments with a transition temperature of 15 ºC was purchased from LCRHallcrest. The one with a transition temperature of 33 ºC was purchased from Karlsson Robotics. The one with a transition temperature of 65 ºC was purchased from Acumind Pigments). A layer of uncured Ecoflex® with a thickness of ca. 0.1 mm was cast atop the pigment mixture layer and placed in a refrigerator set at -10 °C for 4 hours to allow the uncured Ecoflex® to penetrate into the porous spacing between the pigment particles followed by cured at 80 °C for 2 h. The as-prepared thermal mapping unit was then attached to the palmar aspect (front side) of a dummy finger. To prepare the HSSU, the NSSU was pre-stretched to 29% strain and then attached to the PDMS substrate (thickness ≈ 1.3 mm) with silicone adhesive prior to release. To apply for finger motion monitoring, the HSSU was placed right atop the dorsal aspect (back side) of the middle finger joint part and two ends of the HSSU was bond to the non-bendable segments with silicone adhesive.

Characterizations:

The HSSU, NSSU, and LSSU units were cut into rectangles and mounted on a custom-built stretching tool to characterize their optical performance. The reflectance spectra of all of the samples were characterized on a Shimadzu UV-2450 UV-Vis spectrophotometer and/or a Mprobe VIS-NIR thin film measurement system. The transmittance changes of the PEDOT: PSS electrochromic response was recorded on a PerkinElmer ultraviolet/visible/near-infrared Lambda 900 spectrophotometer. The topography of the MSR units were recorded on an AmScope ME 520TA optical microscope. All of the digital photos and videos were captured with an iPhone 6 Plus. The La*b* color coordinates for the MSR units image taken by iPhone 6 Plus were analyzed by the Adobe Photoshop CS4 software to extract the average value.

Definition of the degree of red (DR):

$DR = 100\% \times |a^*_{x\%} - a^*_{0\%}| / |a^*_{300\%} - a^*_{0\%}|$, where $a^*_{0\%}$, $a^*_{x\%}$, and $a^*_{300\%}$ are the a* value of the La*b* coordinates at 0%, x%, and 300% strain, respectively

Definition of the reduction of green (RG):

$DR = 100\% \times (1 - |a^*_0 - a^*_x| / |a^*_0 - a^*_{min}|)$, where a^*_0, a^*_x, and a^*_{min} are the a* value of La*b* coordinate at 0 (s), x (s), and the time to reach minimum a* value, respectively

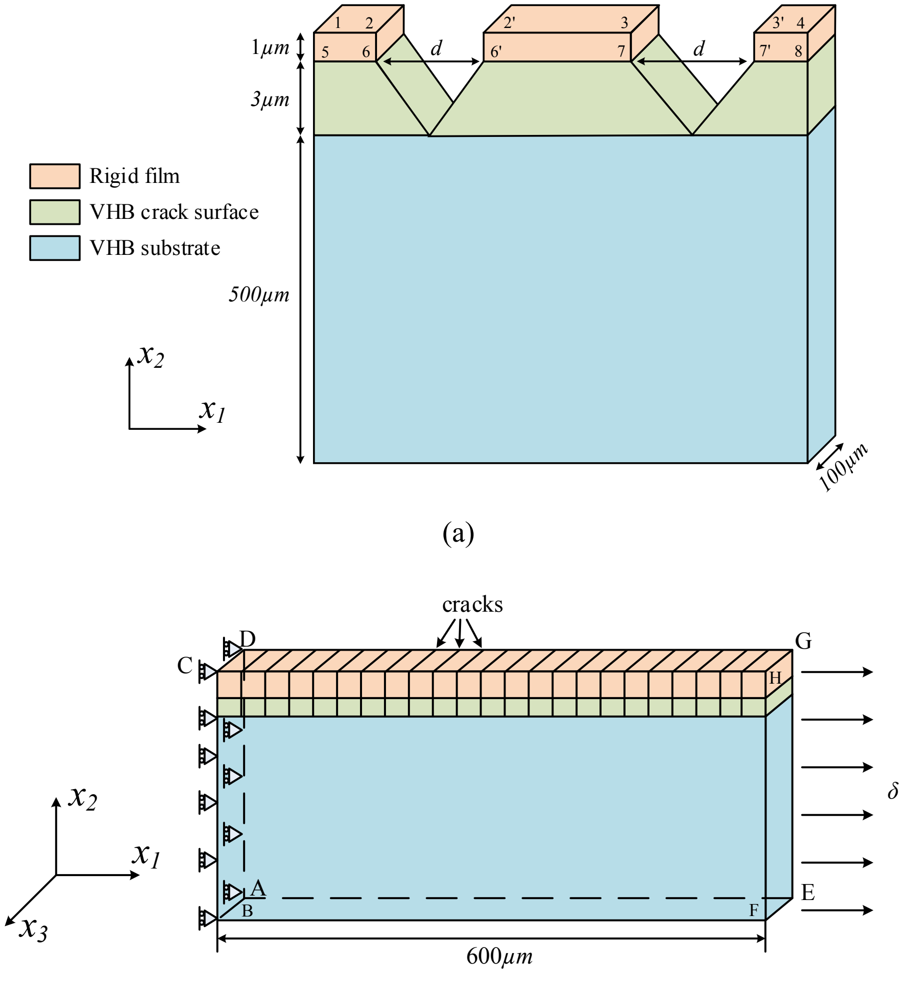
Definition of the degree of violet (DV):

DV= 100%×|b*$_x$ - b*$_0$|/|b*$_{max}$-b*$_0$|, where b*$_0$, b*$_x$, b*$_{max}$ are the a* value of La*b* coordinate at 0 (s), x (s), the time to reach maximum b* value, respectively.

Finite Element (FE) simulation of crack evolution in rigid light shielding layer and VHB:

The crack opening response was simulated using the commercial software ABAQUS (version 2017). The VHB substrate was modeled as an incompressible, hyperelastic material using the Arruda-Boyce model with a ground state shear modulus of 3.2 MPa and a locking parameter (λm) of 1.17[2]. The rigid thin film, modeled as an incompressible elastic solid with a Young's modulus of 10 GPa, was tied onto the substrate by enforcing the displacement continuity at the interface. Both the VHB substrate and the thin film were meshed using 3D hybrid linear elements, C3D8H. The distributed cracks on the thin film, which were developed after the pre-stretch procedure, were modeled as dummy nodes at the crack interface. These cracks were fully opened through the thin film and arrested in the VHB substrate, as schematically shown in Figure S1a. In the present model, 30 parallel cracks were embedded along the loading direction, and the crack depth followed a normal distribution based on the average value and standard deviation measured from the experiment. The crack size during the deformation was determined by averaging the relative displacements of the two dummy nodes at the crack interface, *d* (see Figure S1a), of the 30 cracks.

The boundary conditions for the film-substrate system subjected to uniaxial tension are shown in Figure S1b. A uniform displacement field is prescribed on the surface CDGF ($U_1 = \delta$), while the opposite surface (surface BAHE) is constrained along the x_1-direction ($U_1 = 0$). Additionally, point A is fixed ($U_1 = U_2 = U_3 = 0$) to prevent rigid body motion, and point B is restrained from moving along the x_2-direction ($U_2 = 0$). The rigid film thickness, crack spacing, and crack depth in the VHB substrate are shown in Figure S1a.



(a)

(b)

Figure S1. FE model for crack opening response. (a) Dummy nodes used to represent crack opening. (b) Boundary conditions.

FE simulation of the pre-stretch of NSSU and the re-flattening of HSSU:

The bent and re-flattened responses were simulated using the commercial software ABAQUS (version 2017). The NSSU was pre-stretched to 29% strain under pure tension. The geometry and boundary conditions are shown in Figure S2a. Since the rigid film on the NSSU surface is cracked, the strain energy stored in the rigid film can be neglected in the pre-stretched sample. Therefore, the NSSU can be represented by the VHB layer which is modeled as an incompressible hyperelastic material using the Arruda-Boyce model with a ground state shear modulus of 3.2 MPa and a locking parameter (λm) of 1.17[2]. The stress state of each element in NSSU obtained from the pre-stretch simulation was stored and subsequently passed into the re-flattening simulation.

Because of the stress development in NSSU during the pre-stretch process, once it is attached to the PDMS substrate, the entire unit, which is called as HSSU, tends to bend. In the experiment, the entire unit was compressed on a flat, rigid surface to restore the flatness. To capture this response, the nodal coordinates and the pre-defined stress state of NSSU were imported from the last step of the pre-stretch simulation. Then, NSSU was tied onto the PDMS substrate by enforcing the displacement continuity at the interface. In the FE model, the PDMS substrate was modeled as an incompressible hyperelastic material using the

Arruda-Boyce model with a ground state shear modulus of 15 MPa and a locking parameter (λm) of 1.17[2]. The geometry of the PDMS is shown in Figure S2b. Both NSSU and the PDMS substrate were meshed using 3D hybrid linear elements, C3D8H. The boundary conditions used to re-flatten the unit is shown in Figure S2b.

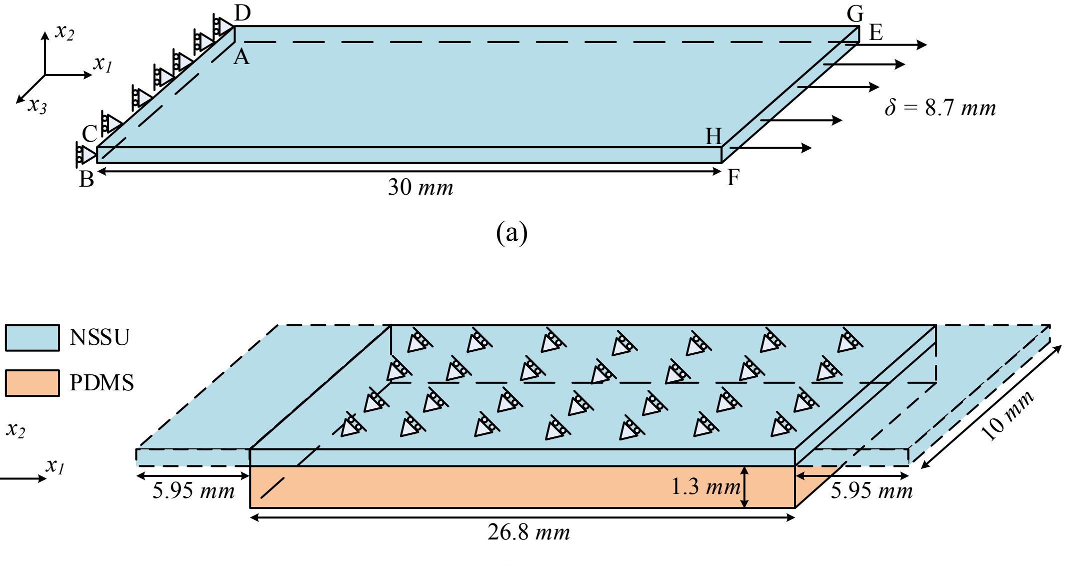


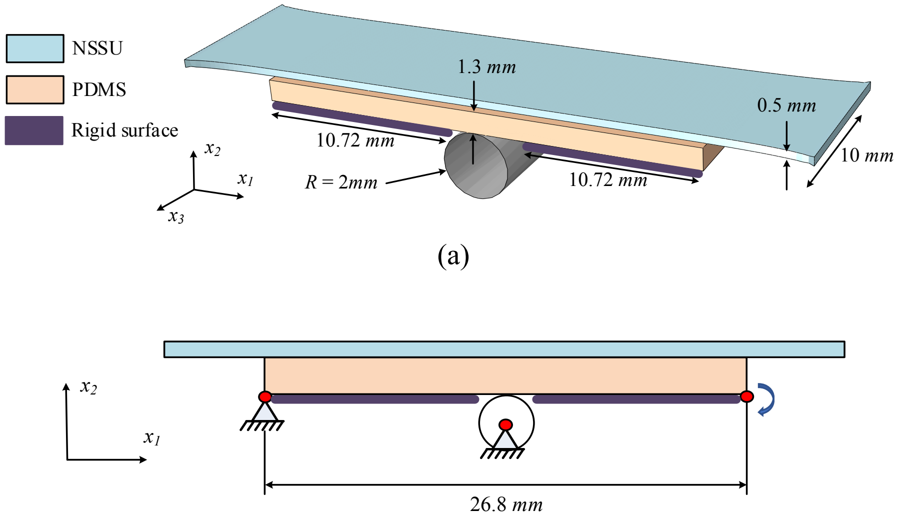
Figure S2. Boundary conditions of the FE models to simulate (a) pre-tension of NSSU and (b) re-flattening of HSSU.

FE simulation of the bending response of HSSU when it is attached on the finger:

First, a similar simulation was carried out to pre-stretch NSSU and re-flatten HSSU due to the stress mismatch of the NSSU and PDMS layers. That is, the nodal coordinates and pre-prescribed stress of NSSU were imported from the last step of the pre-stretch simulation. The NSSU was then tied onto the PDMS substrate by enforcing the displacement continuity at the interface.

To simulate the deformation response of HSSU when it is attached on the finger, the finger motion was captured by applying a bending moment on one edge of the two rigid surfaces that were connected through a roller, as shown in Figure S3. In the simulation, the rigid surfaces were attached at the bottom of the PDMS substrate. The rigid roller was assigned right under the middle of the PDMS substrate to represent the finger joint. Reference points were attached on the rigid pieces to apply the boundary conditions to capture the finger movement. The geometry and locations of the reference points are shown in Figure S3a.

In order to simulate the finger bending motion in various angles, displacements and rotations were assigned at the reference points to capture the motion of finger. The reference point attached to the roller was fixed. The reference point attached on one of the rigid surfaces was fixed, while various rotation angles were prescribed on the other reference point attached on the rigid surface. Detail of the boundary conditions areshown in Figure S3b.



(a)

(b)

Figure S3. (a) 3D FE model to capture the deformation response of HSSU when it was attached on a finger. (b) 2D view to show the boundary conditions.

Smartphone colorimetric analyzing system for monitoring finger bending:

An Android application for smartphone colorimetric analysis was developed by Android Studio independent development environment (IDE). The general method for measuring the degree of finger bending was realized by reading the color change on the middle free part of HSSU atop the middle finger joint (This app only focuses on calculating the finger bending degree under (Vis, <33^{o}C) condition). This smartphone application provides an easy-to-use and portable method to monitor the degree of finger bending.

The application has 3 main functions which are "taking pictures", "reading La*b* value from image" and "calculating the bending degree". The application uses the camera that comes with any smartphone to take pictures for analysis. For reading La*b* value color, this application uses the bitmap class in Android platform, which can read the RGB value of a pixel from the image and then convert this RGB value to a La*b* value via the ColorUtils class in android support library. To improve the accuracy of reading the image color, the application provides an adjustable black square box which can read all pixels inside a square area and calculate the average RGB value, followed by converting to an average La*b* value. The size of the black square box can be changed by the "-",''+" button and its position can be moved by the touch screen.

With these functions, the application works as follows to calculate the finger bending degree:

In the first step, the button "take a picture" is pressed to take a picture of middle free part of the HSSU on a flattened finger at 0 bending degree and then the "set up calibration" button is clicked to read La*b* value for a finger with 0 bending degree. The black square box is used to select the image of middle free part of the HSSU atop the middle finger joint. The app converts the selected image area into an average La*b* value (Denoted as L_o, a^*_o, b^*_o) for calibration.

In the second step, the finger is bent to an arbitrary degree under the same lighting environment and the " take a picture" button is again pressed, followed by clicking the "calculate bending degree" button. Selecting the middle free part of HSSU via the black square box. The app then converts the selected image area into another new average La^*b^*(Denoted as L_x, a^*_x, b^*_x) value. The system automatically calculates the a^*value change to 0 bending degree (= a^*_x- a^*_o) and inputs this value into a pre-set calibration curve, which is used to identify the variation of finger bending degree to different a^* value change to 0 bending degree, thus calculating the bending degree. With the result, the calculated finger bending degree is displayed on the screen.

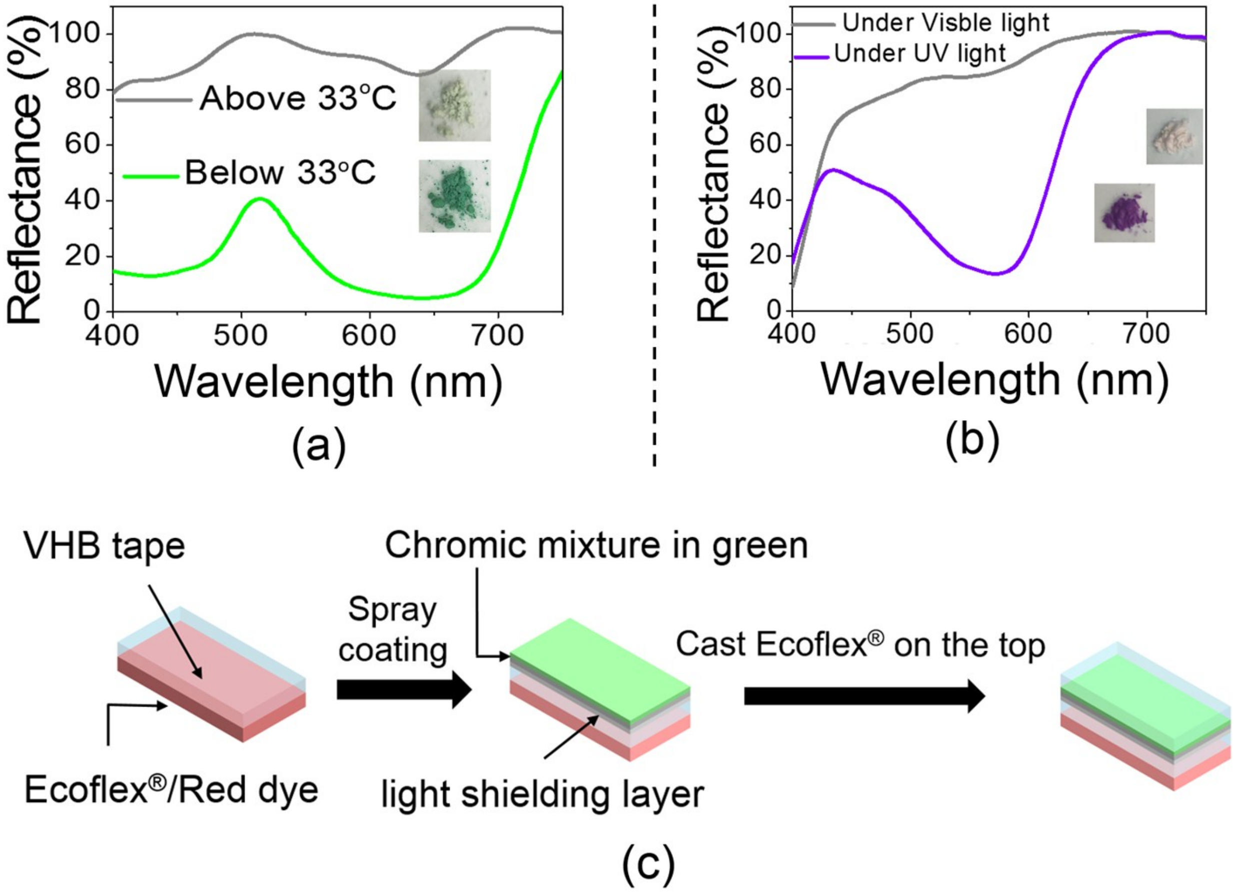


Figure S4. Reflectance spectra, chemical formula, and digital photos of (a) thermochromic and (b) photochromic pigments and (c) the fabrication scheme of the NSSU.

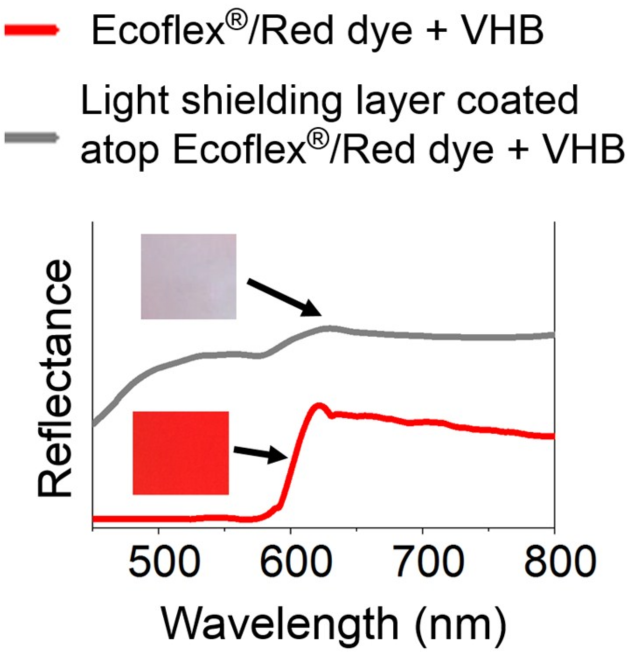
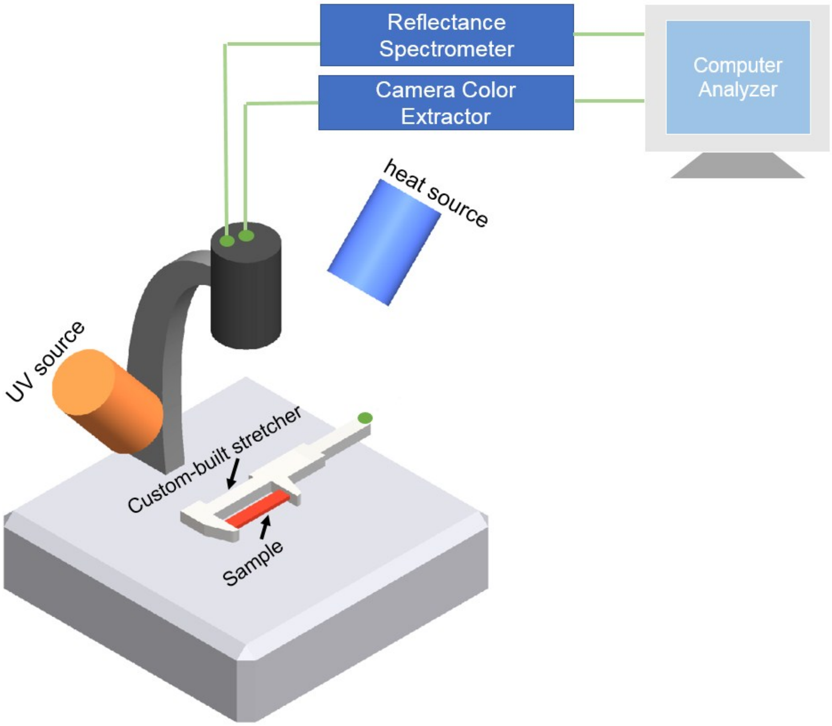
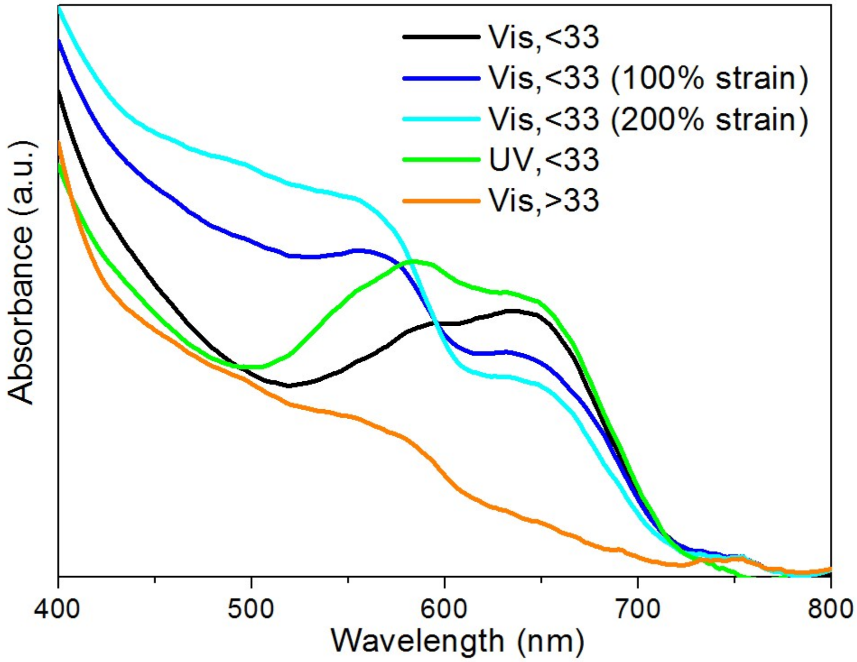


Figure S5. Reflectance spectra and digital photos showing that the addition of the light shielding layer can effectively block the red color substrate



(a)



(b)

Figure S6. (a) Set-up for the characterization of the MSR units. (b) The absorbance spectra of the MSR performance for the NSSU

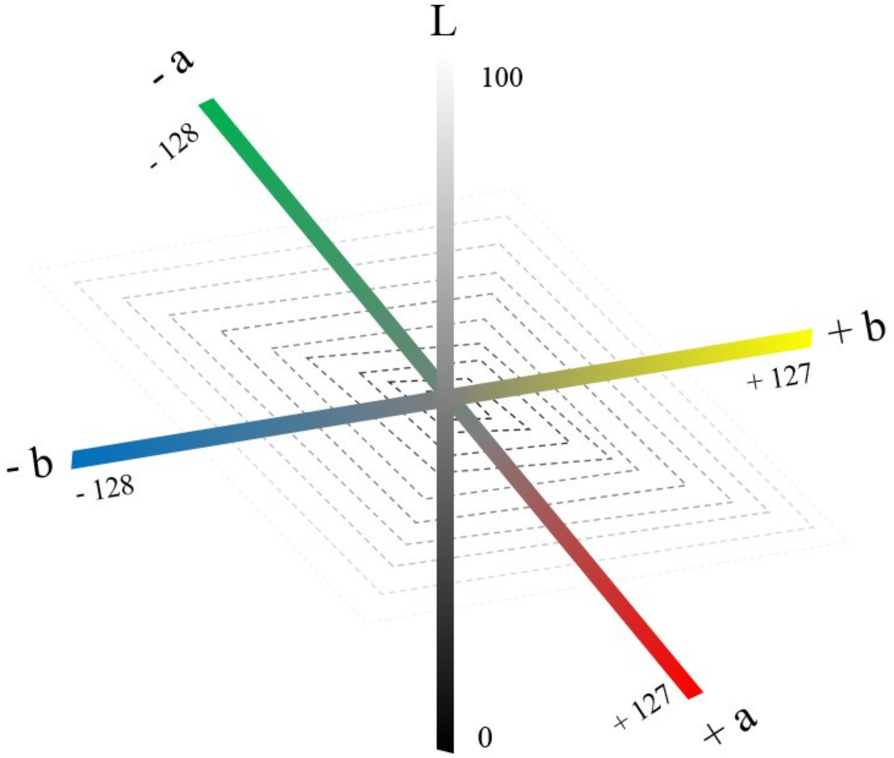


Figure S7. Definition of the La*b* color coordinate. Please see the following link for details: https://en.wikipedia.org/wiki/CIELAB_color_space.

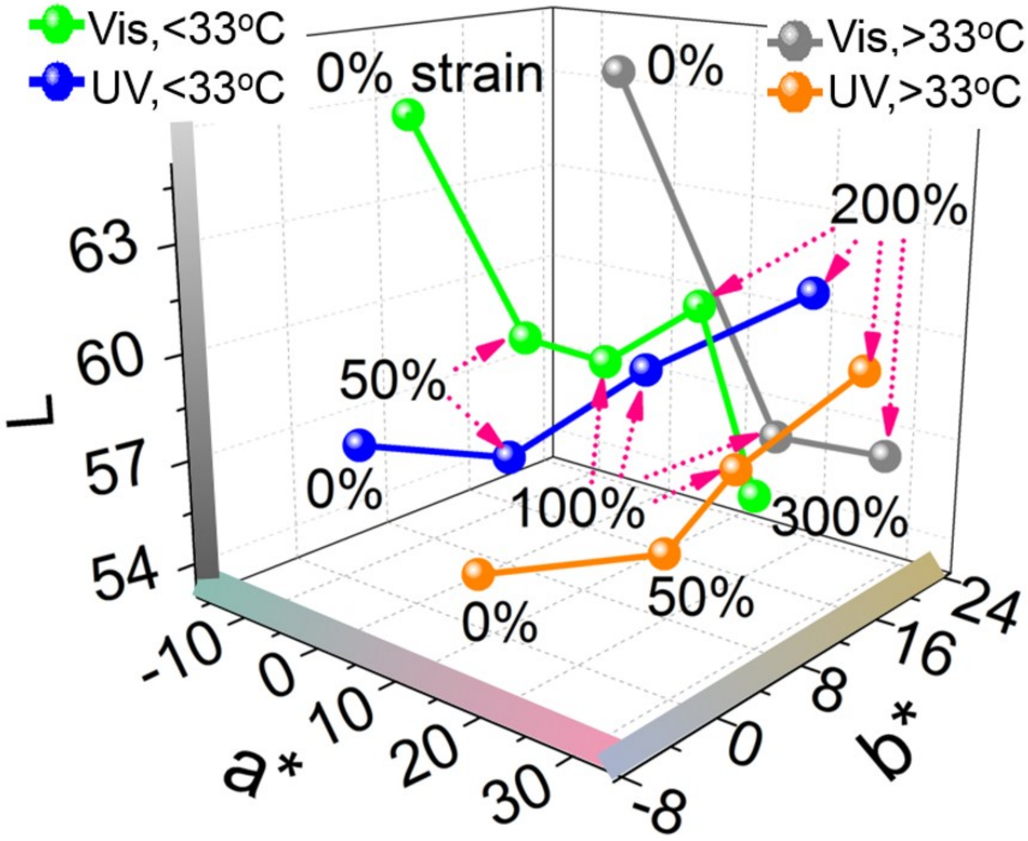


Figure S8. La*b* color coordinates for the mechanochromic effect of the NSSU units under (Vis, <33 ºC), (Vis, >33 ºC), (UV, <33 ºC) and (UV, >33 ºC) conditions.

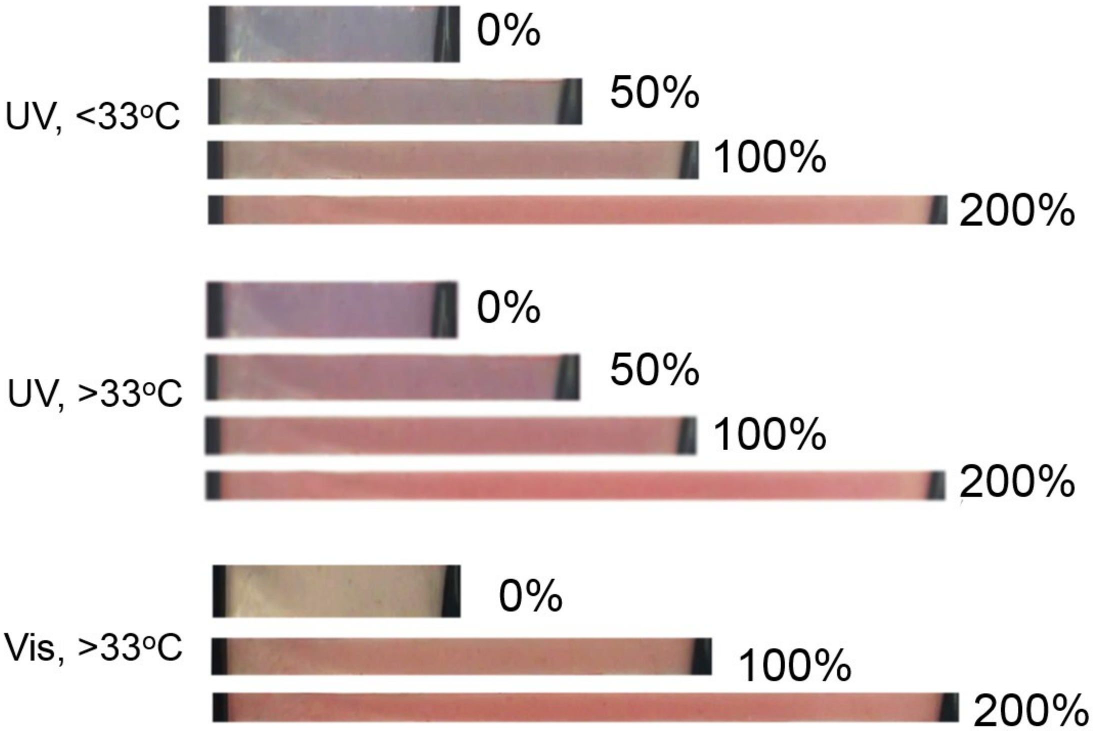


Figure S9. Digital photos for the mechanochromic effect of the NSSU units under (Vis, <33 ºC), (Vis, >33 ºC), (UV, <33 ºC) and (UV, >33 ºC) conditions.

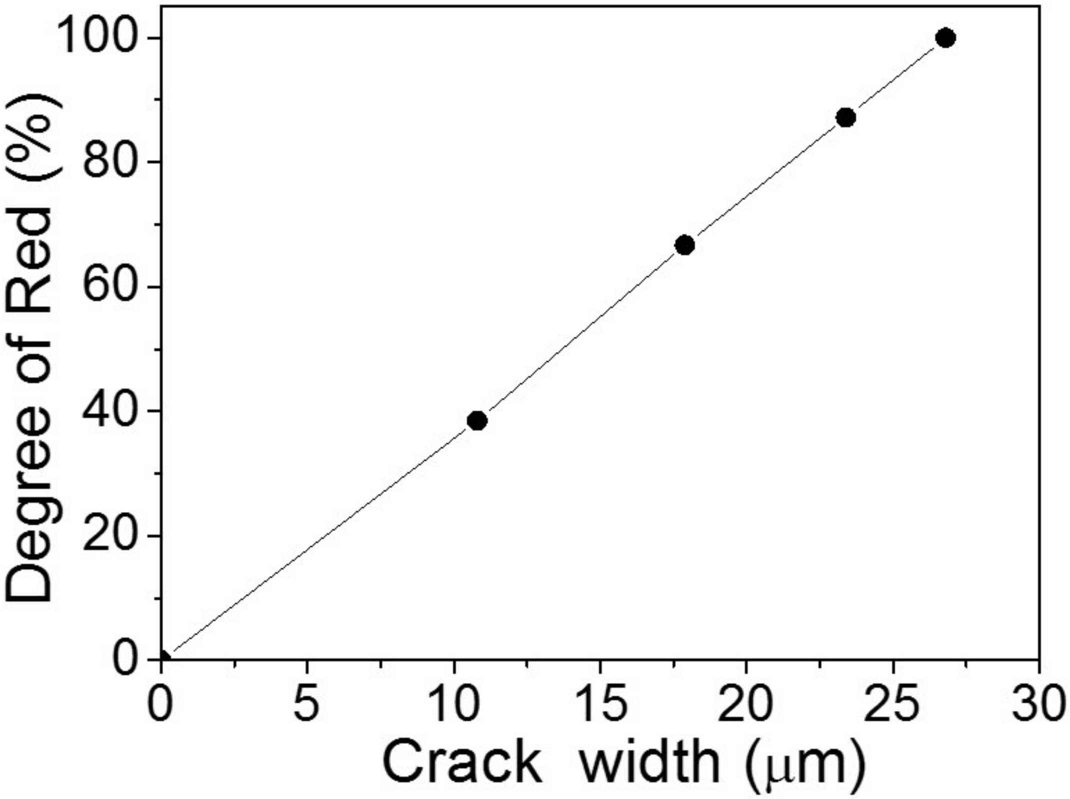


Figure S10. Degree of red as a function of crack width for the NSSU sample.

Table S1. Time for the NSSU unit to reach maximum degree of violet (DV) under different UV output power intensity.

UV intensity (µW/cm^2)	Time to reach maximum degree of violet (DV) (s)
536	7.0
438	7.6
325	11.7
223	13.3
139	18.2
59	18.3

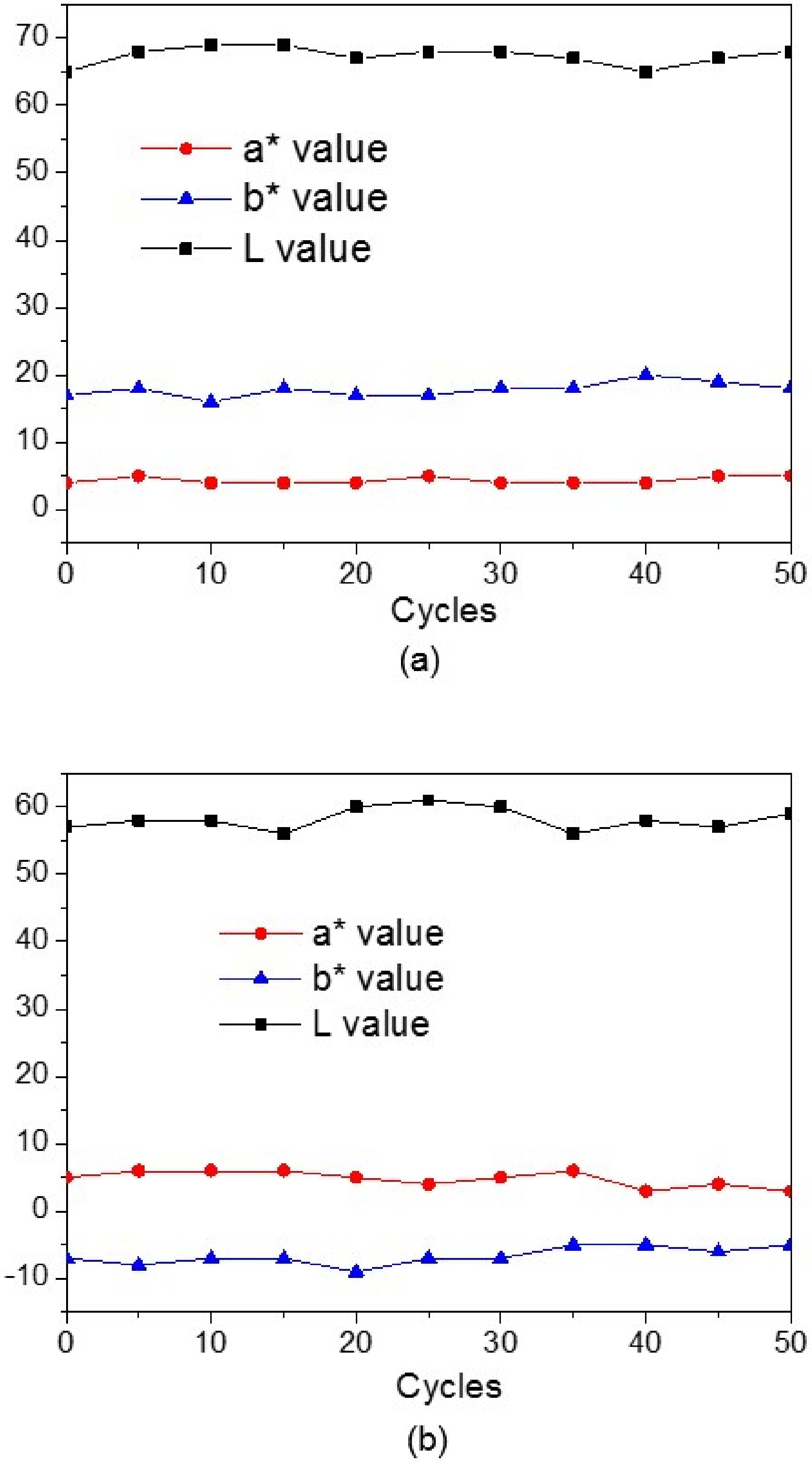


Figure S11. La*b* coordinates variations for (a) thermochromic (testing conditions: heated to 40 °C) and (b) photochromic (testing conditions: exposed to 536 μW/cm^2 UV for 15 s) responses for the NSSU unit during the cyclic test.

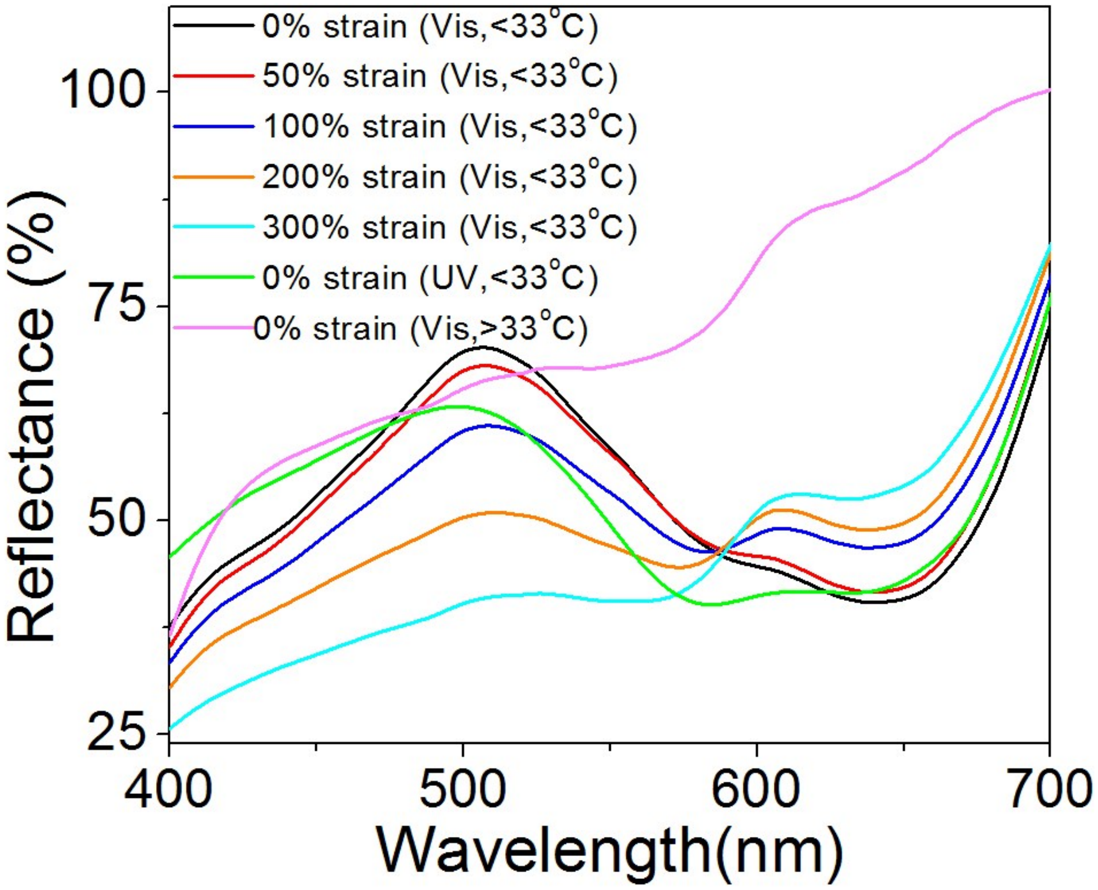
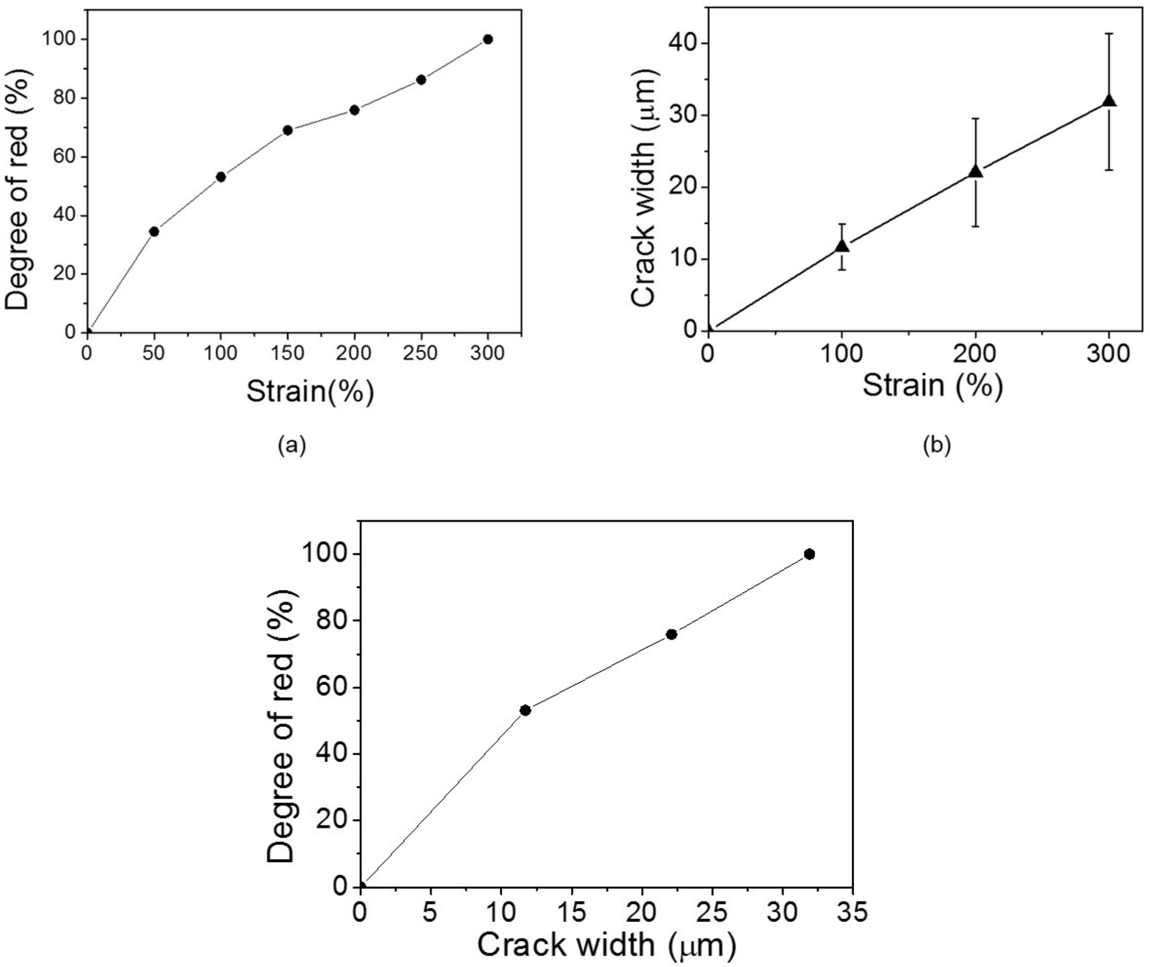


Figure S12. Reflectance spectra of MSR performance of the LSSU unit.



(a)

(b)

(c)

Figure S13. Strain evolution of (a) the degree of red and (b) the crack width and (c) the degree of red as a function of crack width for LSSU unit.

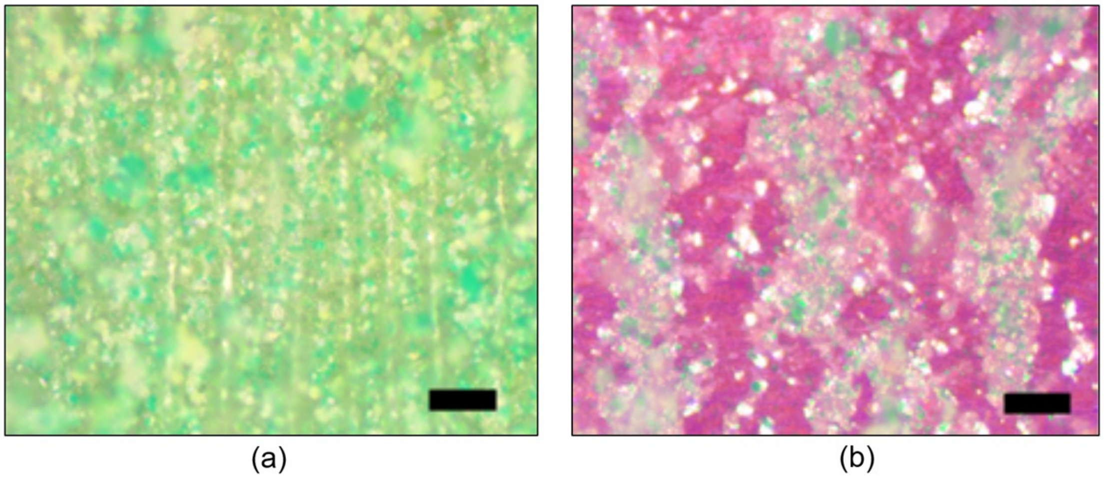

(a) (b)

Figure S14. Optical microscopic images of the topography of LSSU under (a) 0% strain and (b) 300% strain (scale bar = 50 μm).

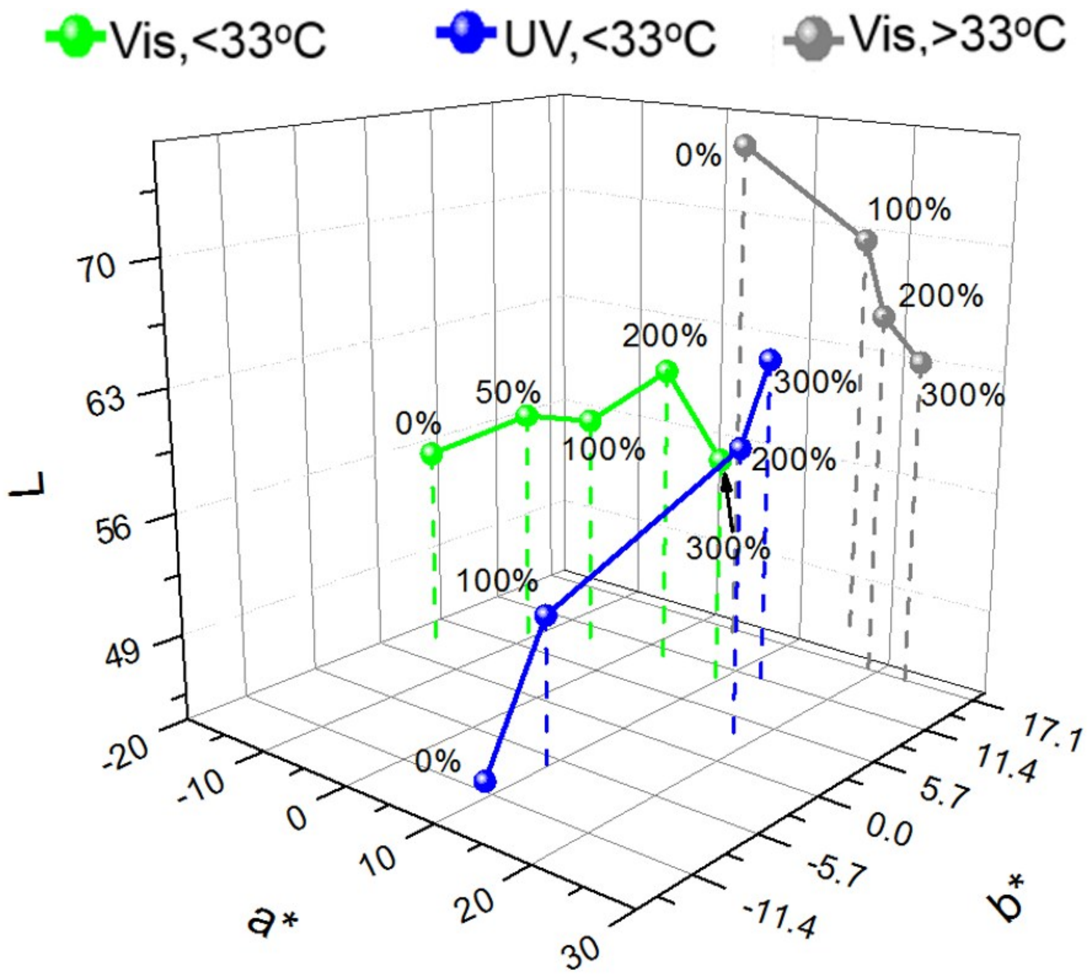


Figure S15. La*b* color coordinates for mechanochromic behavior of the LSSU unit under (Vis, <33 ºC), (UV, <33 ºC) and (Vis, >33 ºC).

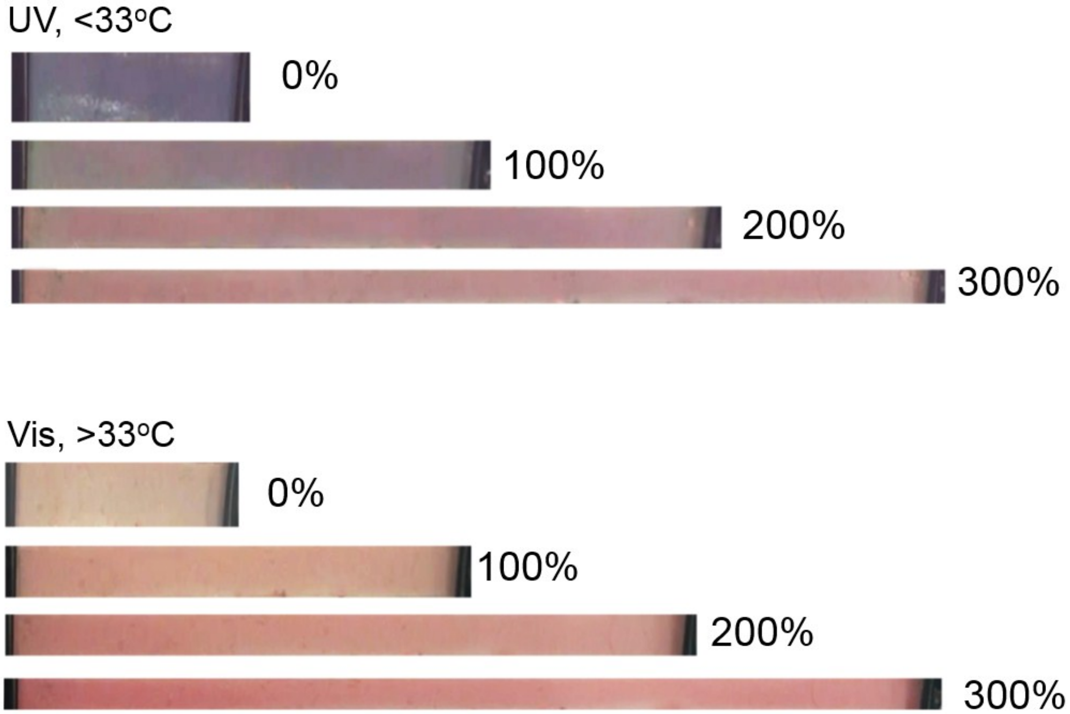
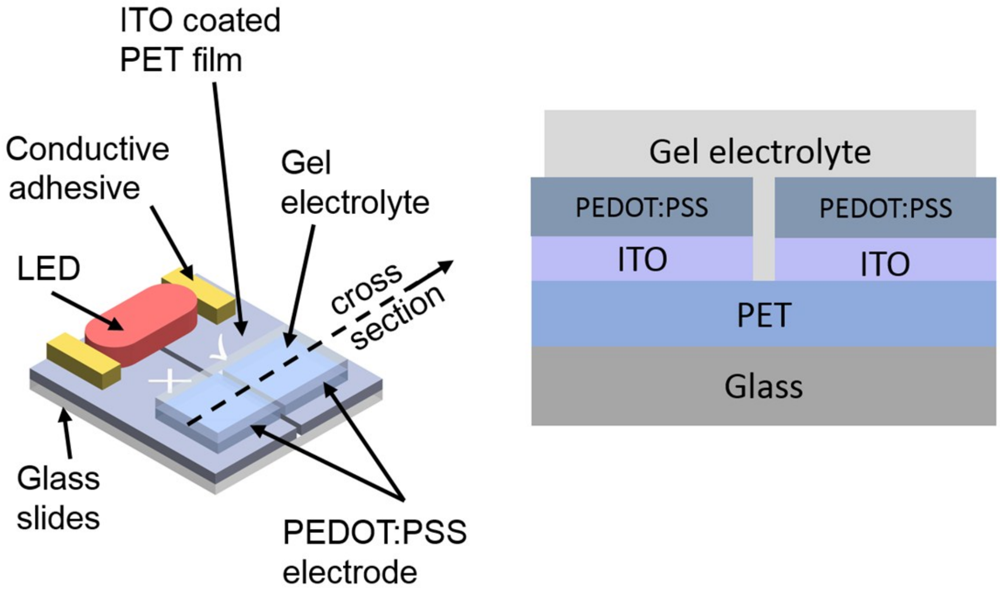
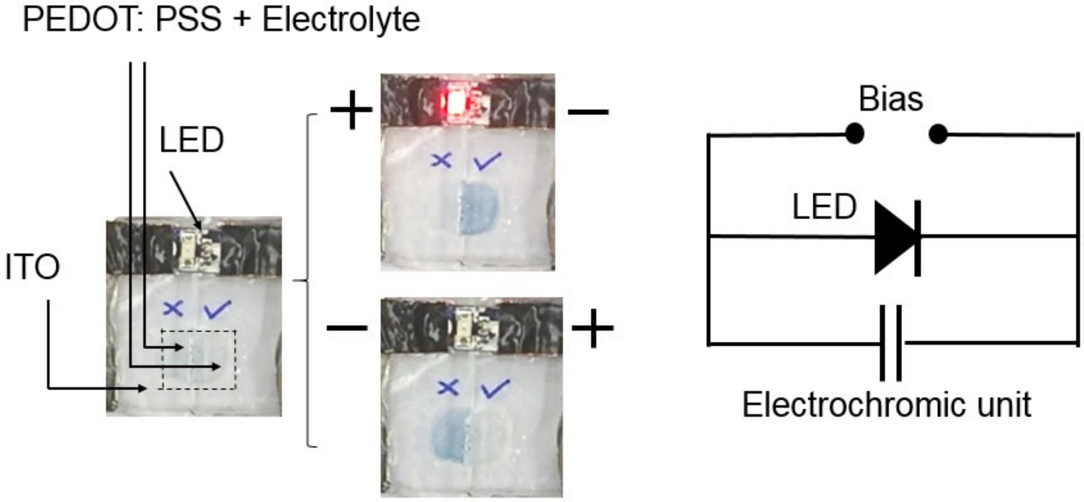


Figure S16. Digital photos for mechanochromic behavior of the LSSU unit under (UV, <33 ºC) and (Vis, >33 ºC).



(a)



(b)

Figure S17. (a) Schematic design of the LED set connected in parallel with an electrochromic unit for stretchable interactive electronics. (b) Circuit layout and performance of a bias direction dependent electrochromic unit connected to an LED set device.

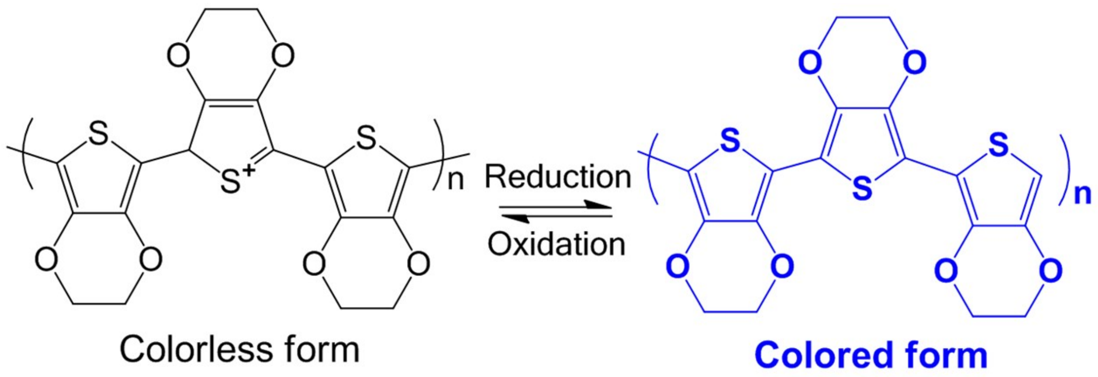


Figure S18. Chemical structure change of the PEDOT: PSS during the electrochromic redox reaction.

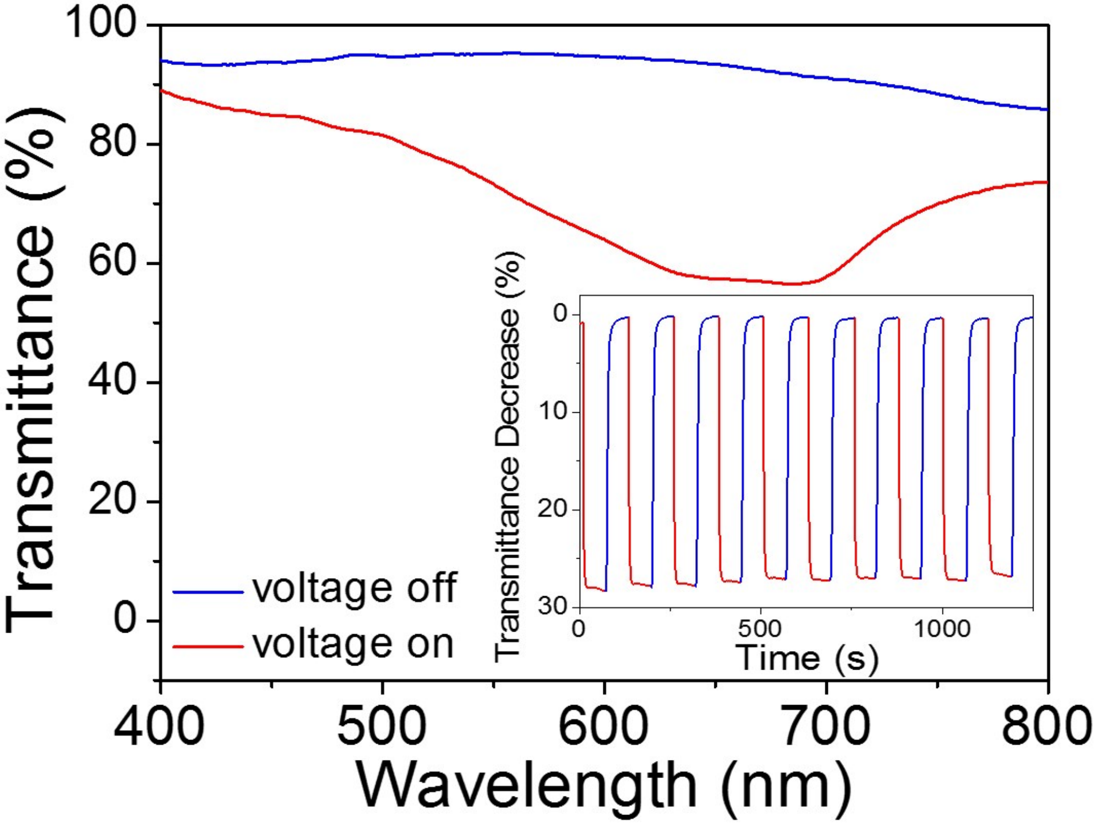


Figure S19. UV-Vis transmittance spectra of the electrochromic performance of PEDOT: PSS to a 2 V bias voltage and the corresponding cycle test.

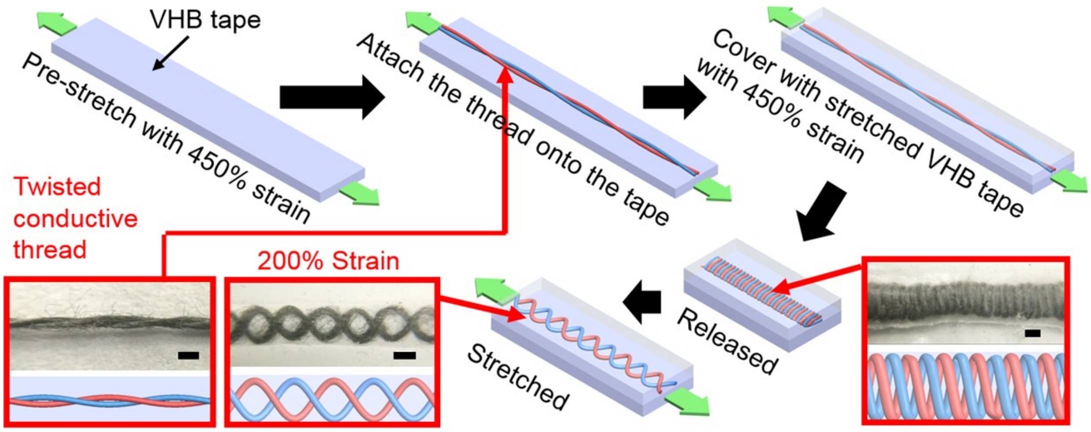


Figure S20. Schemes for preparing a highly stretchable conductor (scale bar = 1 mm) for stretchable interactive electronics.

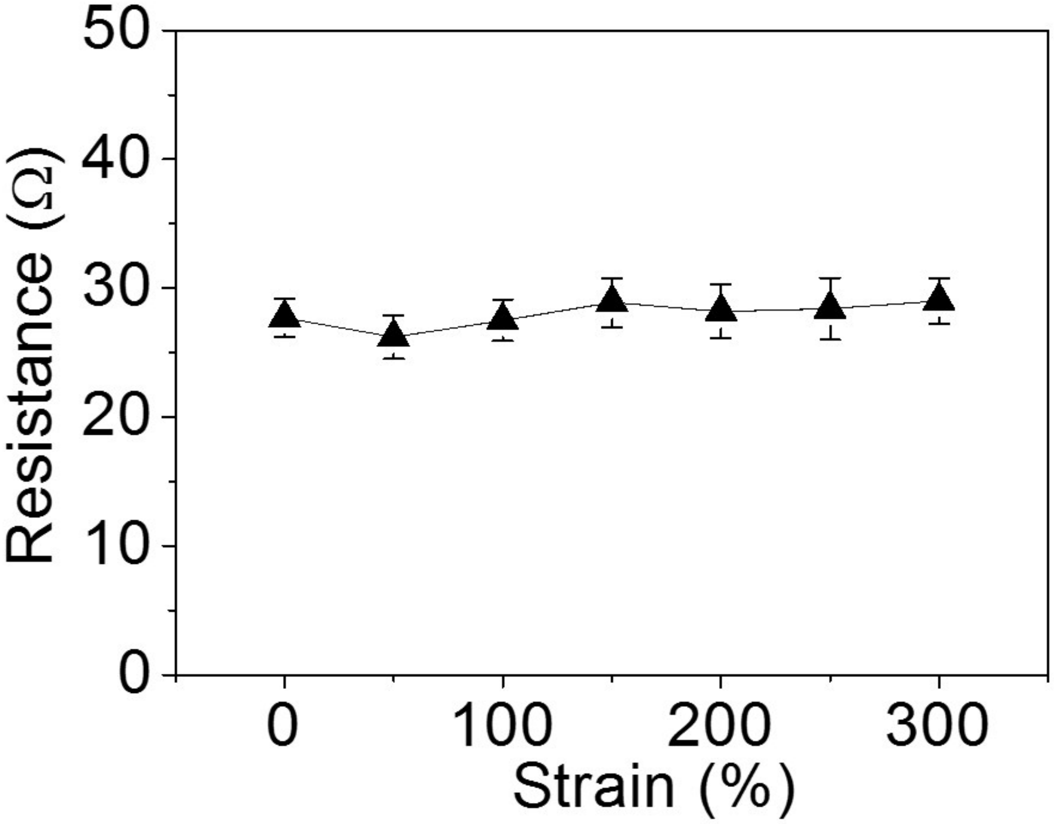


Figure S21. Resistance changes of the stretchable conductor as a function of strain; error bars are defined as standard deviation.

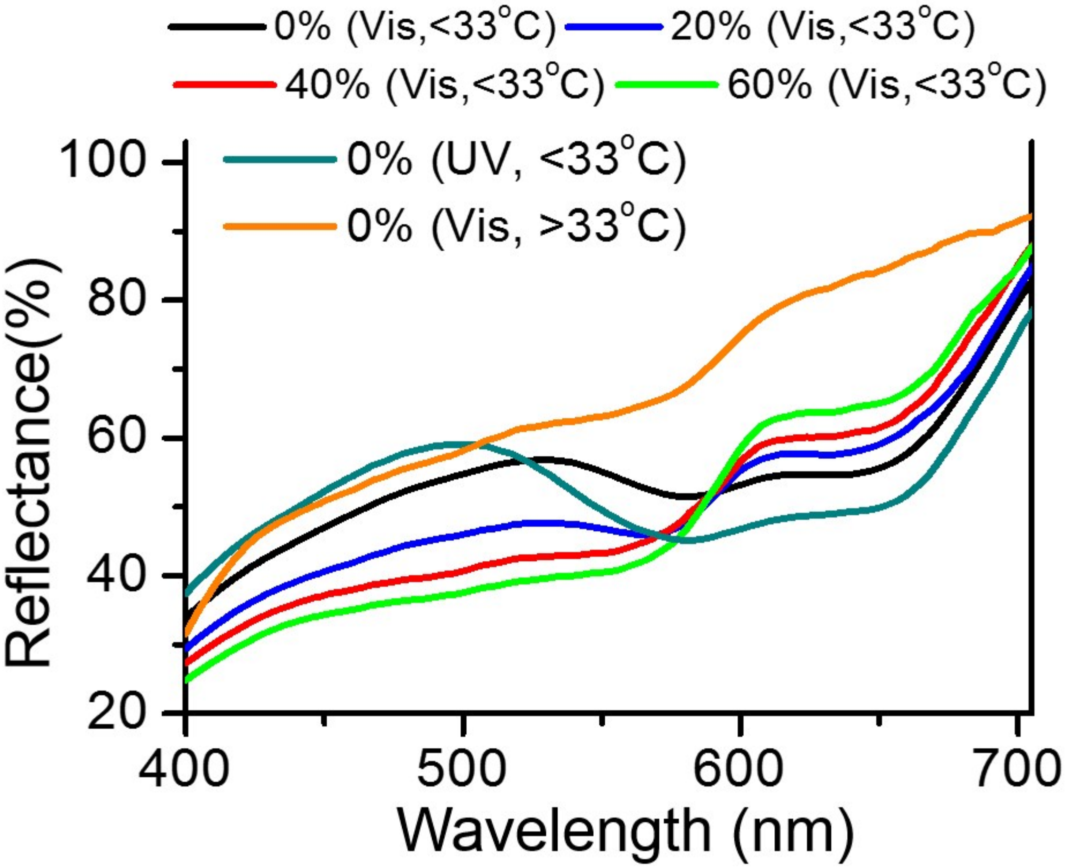


Figure S22. Reflectance spectra of MSR performance of the HSSU unit.

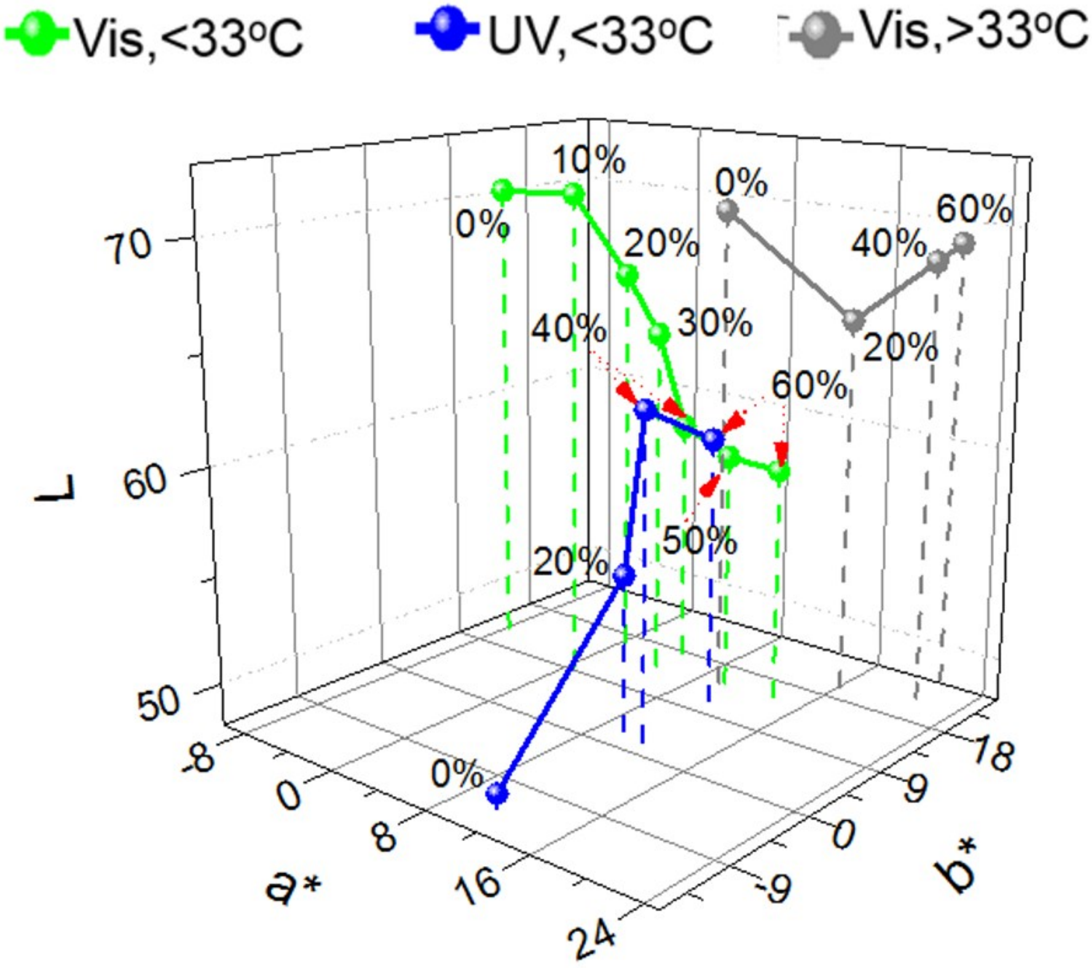


Figure S23. La*b* color coordinates for mechanochromic behavior of the HSSU unit under (Vis, <33 ºC), (UV, <33 ºC) and (Vis, >33 ºC).

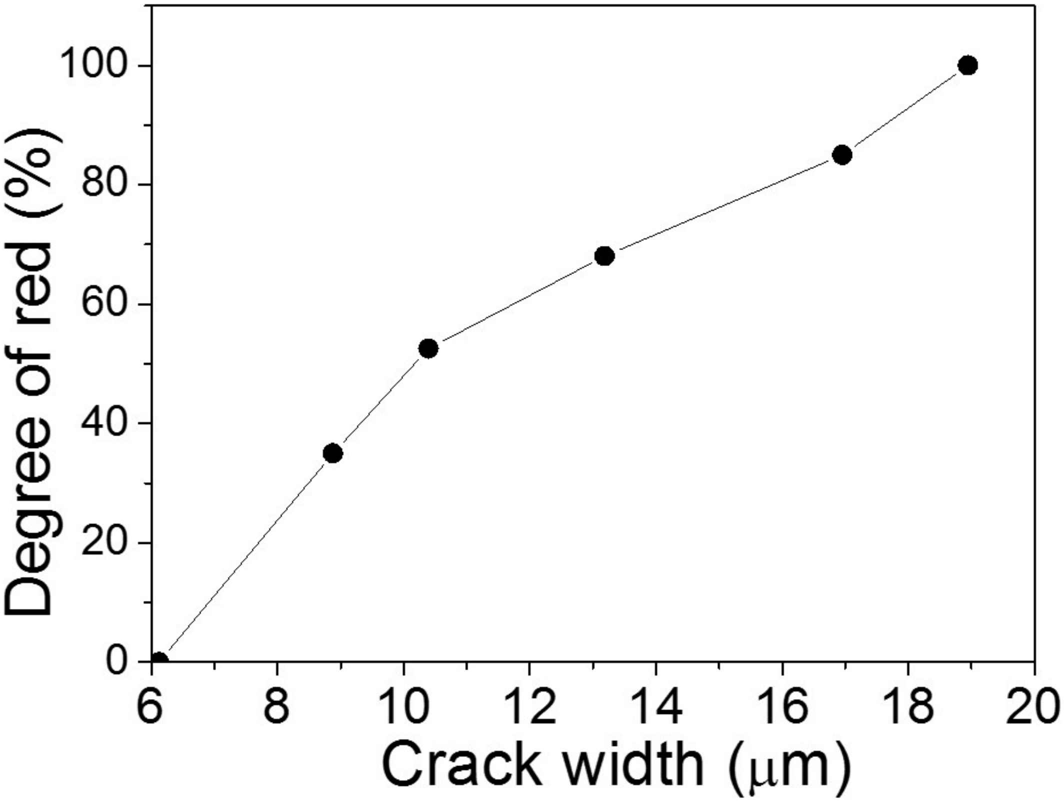


Figure S24. Degree of red as a function of crack width of the LSSU unit.

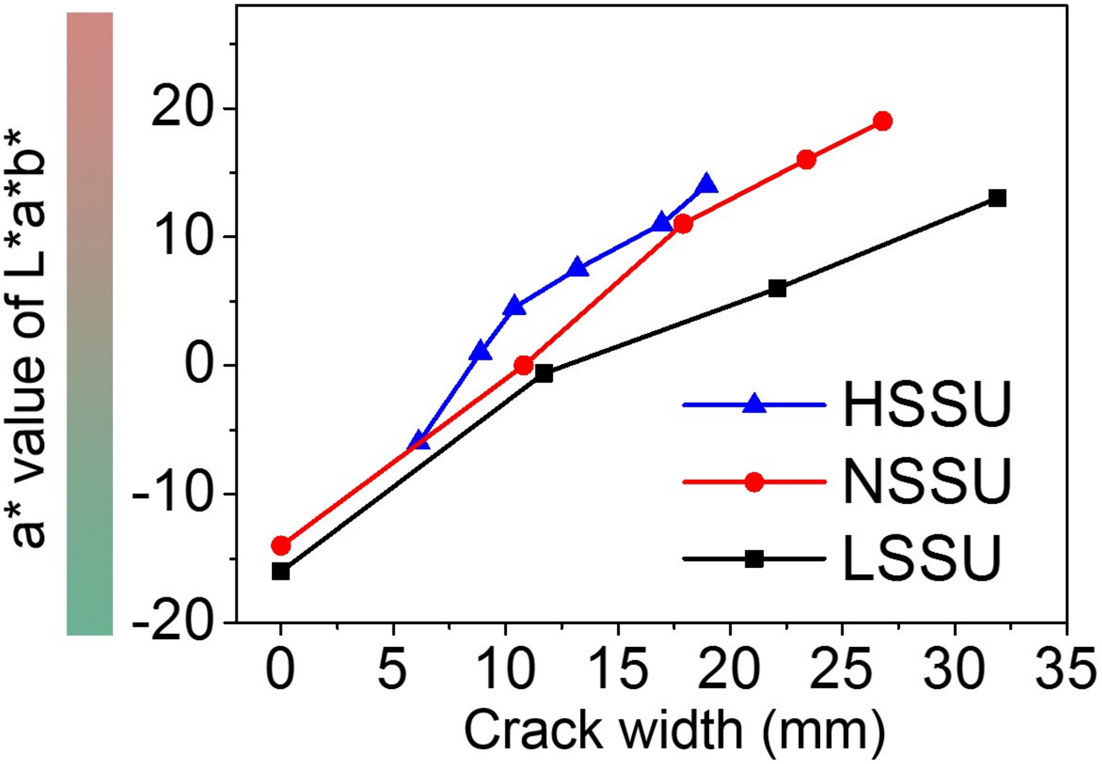


Figure S25. Comparison of evolution of a* value in La*b* coordinates with crack width for HSSU, NSSU and LSSU.

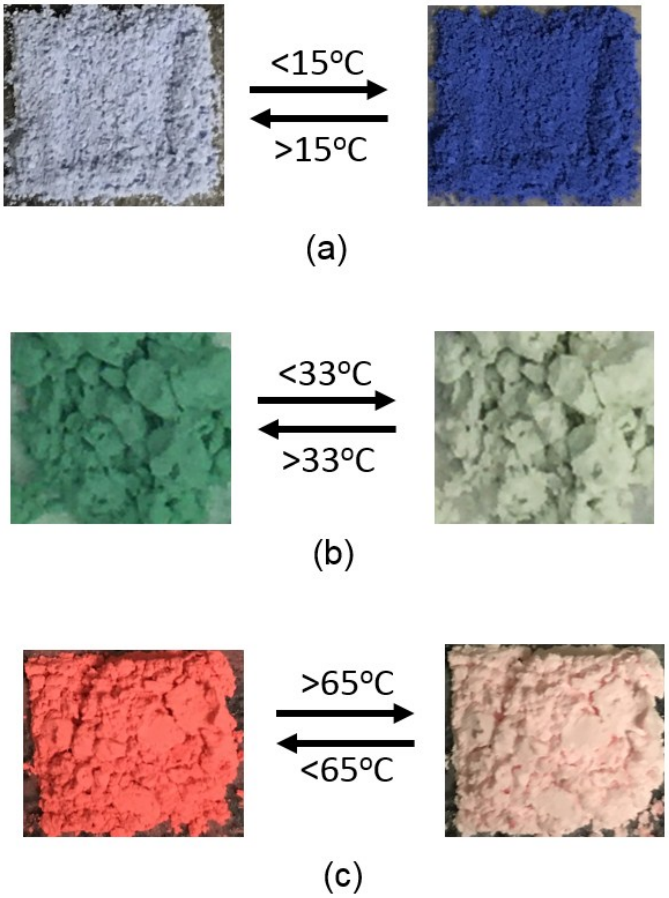


Figure S26. Digital photos of the three thermochromic pigments used in the thermal mapping device with (a) 15 ºC, (b) 33 ºC, and (c) 65 ºC critical transition temperatures.

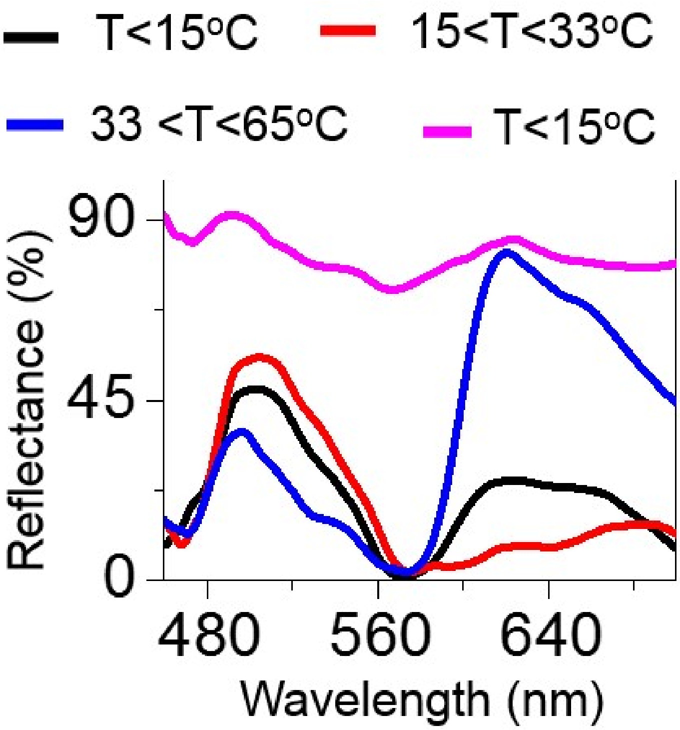


Figure S27. Reflectance spectra of the thermochromic response to different temperature range of the as-prepared thermal mapping unit.

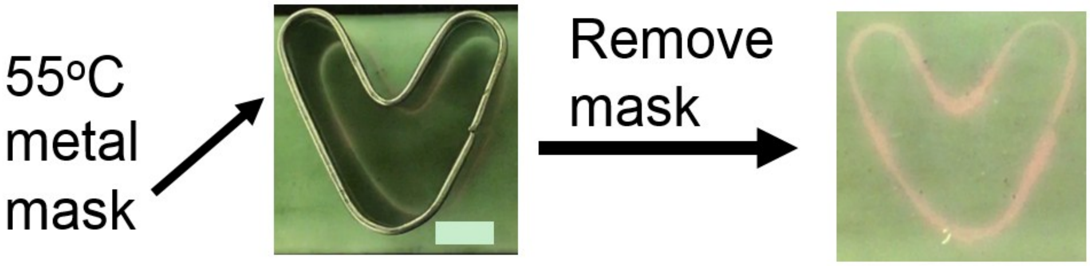


Figure S28. The thermal mapping unit can show precious color pattern upon the removal of a 55 °C metal mask (scale bar = 5mm).

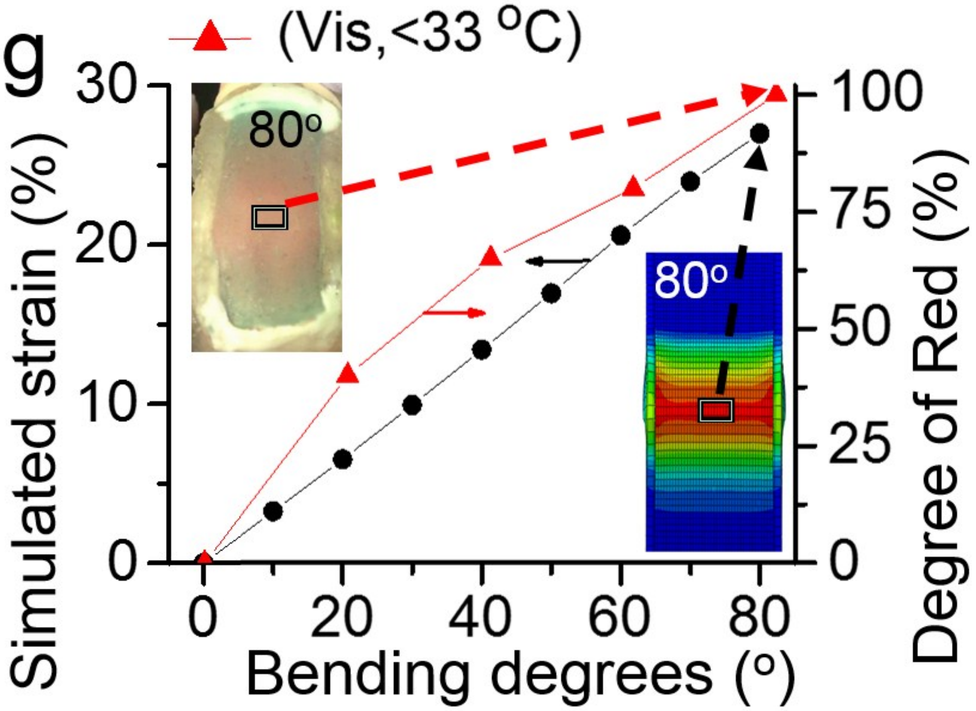


Figure S29. Evolution of the simulated strain level and the experimental degree of red on the middle part of the HSSU on WMEM with different bending degrees.

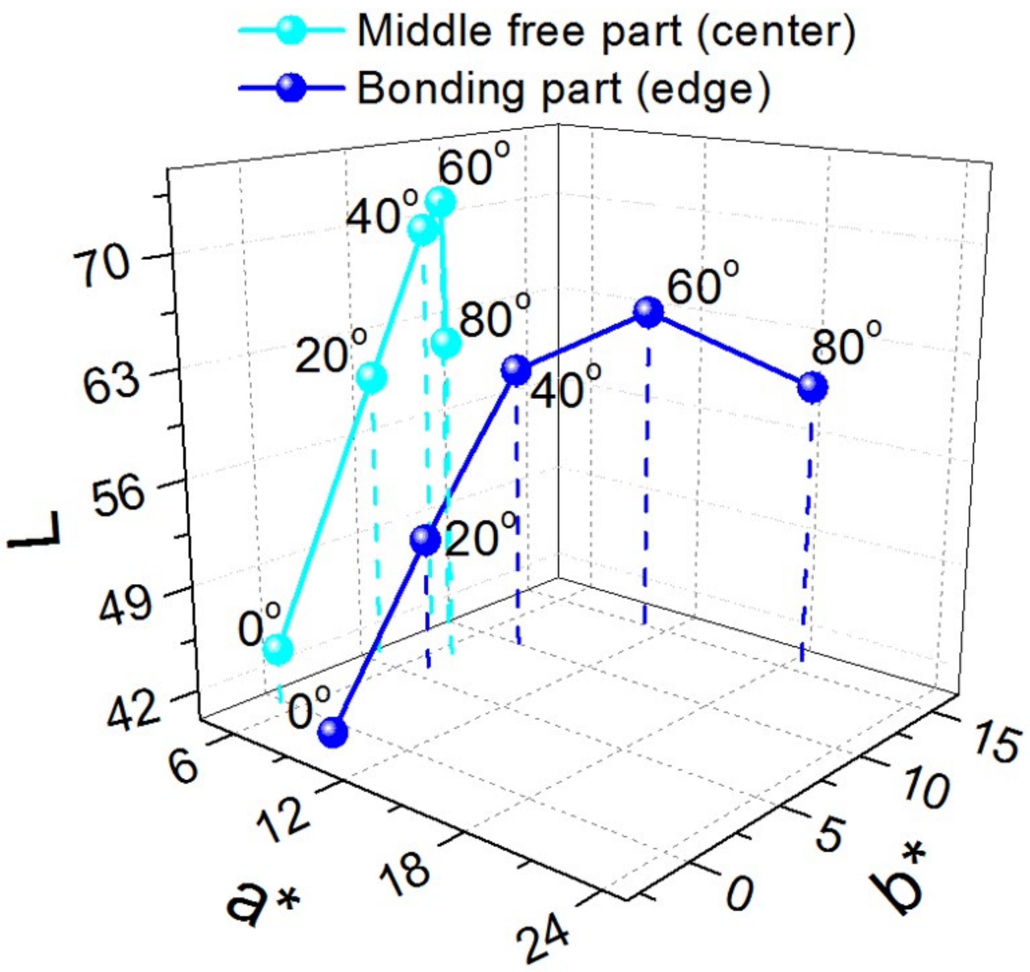


Figure S30. La*b* coordinates of the HSSU on WMEM as a function of bending degree under (UV, <33 ºC).

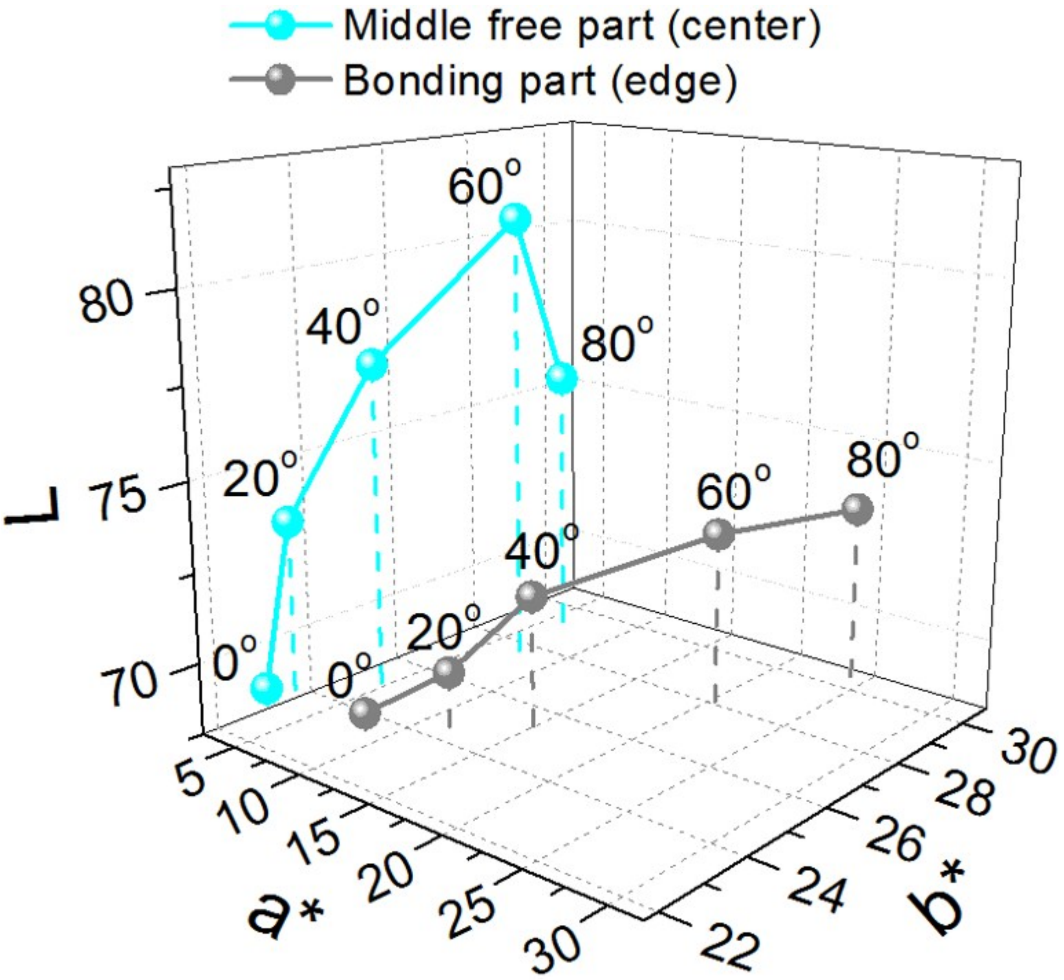


Figure S31. La*b* coordinates of the HSSU on WMEM as a function of bending degree under (Vis, >33 ºC).

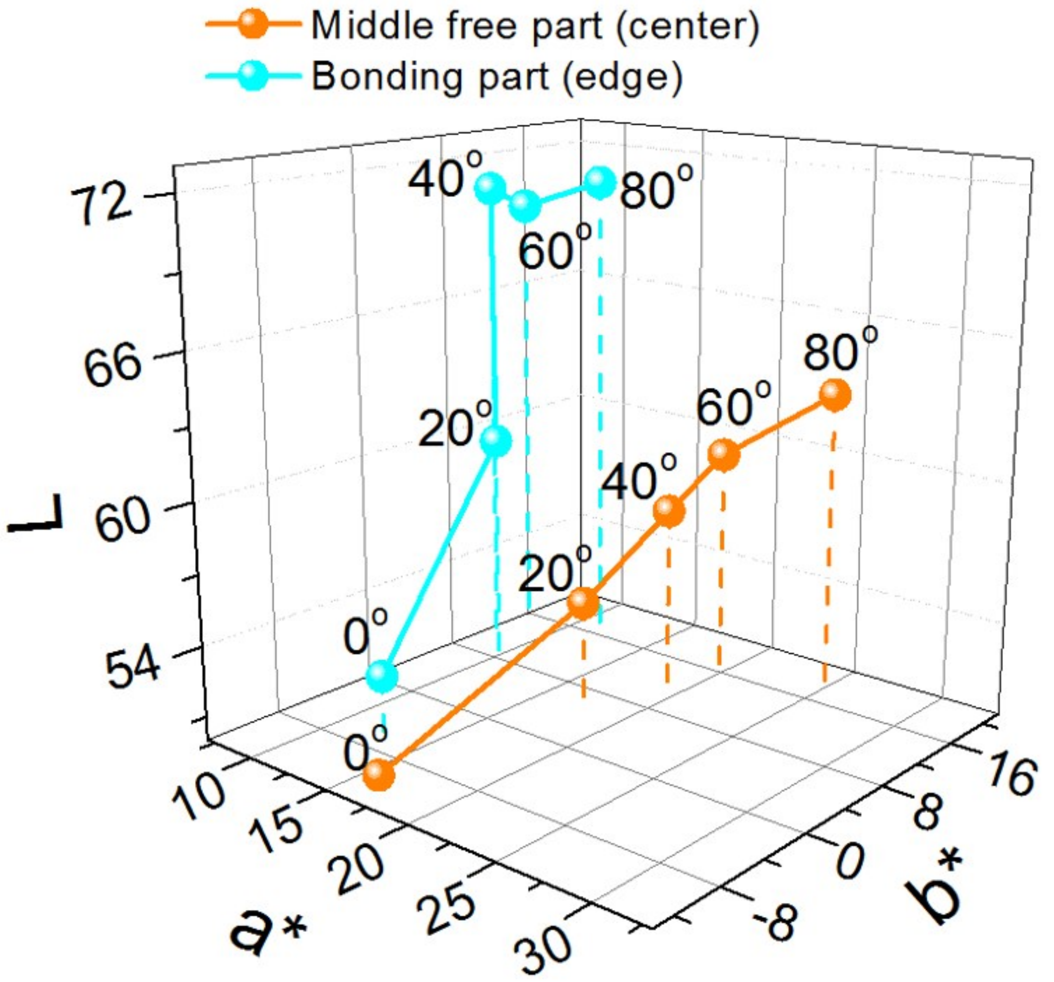


Figure S32. La*b* coordinates of the HSSU on WMEM as a function of bending degree under (UV, >33 ºC).

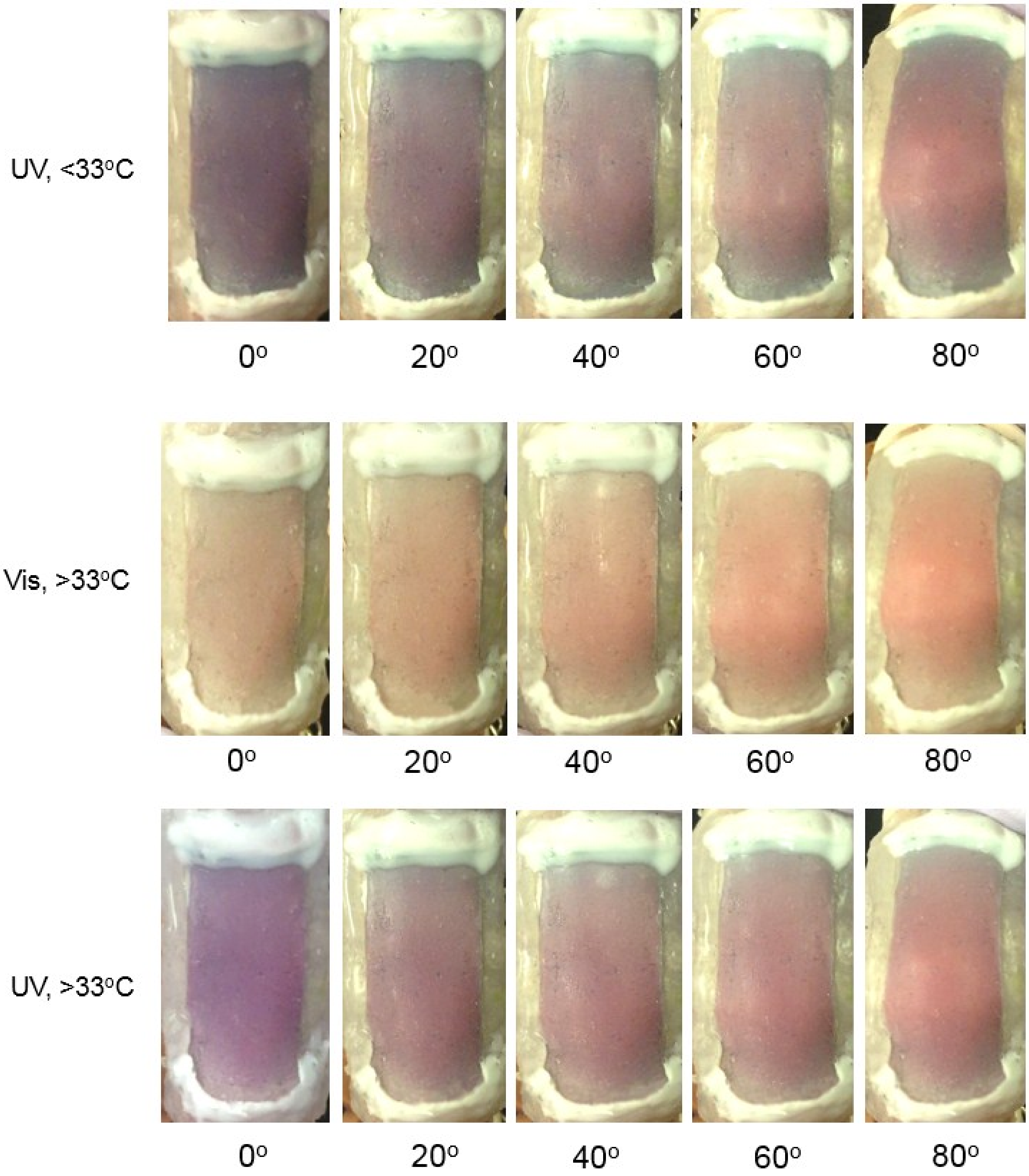


Figure S33. Digital photos of the HSSU on WMEM as a function of bending degree under (UV, <33 ºC), (Vis, >33 ºC) and (UV, >33 ºC) conditions.

Movie captions:

Movie S1. Thermo-/photo-/electrochromic responses of the stretchable interactive electronics.

Movie S2. Mechanochromic response of the stretchable interactive electronics.

Movie S3. Performance of the thermal mapping unit.

Movie S4. MSR performance of the HSSU on monitoring finger bending and environmental conditions.

Movie S5. Smartphone app monitors the finger bending motion through reading the color change of the HSSU.